

Article

Mechanical Properties of Direct Composite Resins and CAD/CAM Composite Blocks

João Carlos Ramos ^{1,*}, Alfredo Marinho ², Ana Messias ^{3,4}, Gabriela Almeida ², Alexandra Vinagre ¹
and Ricardo Dias ³

¹ Institute of Operative Dentistry, Faculty of Medicine, University of Coimbra, 3000-075 Coimbra, Portugal; avinagre@fmed.uc.pt

² Dentistry Department, Faculty of Medicine, University of Coimbra, 3000-075 Coimbra, Portugal; alfredomarinho1996@hotmail.com (A.M.); gabriela.almeida00@gmail.com (G.A.)

³ Institute of Oral Implantology and Prosthodontics, Faculty of Medicine, University of Coimbra, 3000-075 Coimbra, Portugal; ana.messias@uc.pt (A.M.); rbdias@fmed.uc.pt (R.D.)

⁴ Center of Mechanical Engineering Materials and Processes (CEMMPRE), Departamento de Engenharia Mecânica, University of Coimbra, 3030-788 Coimbra, Portugal

* Correspondence: jcramos@fmed.uc.pt

Abstract: The widespread application of CAD/CAM technology in contemporary dentistry led to the development of promising restorative materials, such as resin composite blocks (RCBs). Thus, the present study aims to evaluate the mechanical properties of RCBs, comparing this material to the direct composite resin from the same manufacturer. Samples retrieved from three CAD/CAM resin composite blocks (Tetric CAD (TC), Ivoclar Vivadent, Grandio blocs (GB), VOCO GmbH and Brilliant Crios (BC), Coltene/Whaledent) and four direct composite resins (Tetric EvoCeram (TEC), Ivoclar Vivadent, GrandioSO (GS), VOCO GmbH, Brilliant EverGlow Translucent (BET) and Universal Shade (BEU), Coltene/Whaledent) were submitted to three-point bending flexural test and Vickers microhardness test. The resulting data of the flexural strength were analyzed using one-way ANOVA considering Bonferroni correction for post hoc tests ($\alpha = 0.05$). The flexural modulus and Vickers microhardness results were analyzed using Welch's ANOVA considering Games–Howell correction for post hoc tests ($\alpha = 0.05$). Regarding results, flexural strength and flexural modulus values ranged from 81.1 MPa (BEU) to 246.5 MPa (GB) and 10.6 GPa (BEU) to 20.3 GPa (GB), respectively. GS (121.2) and GB (136.2) groups were associated with the highest microhardness values. According to the post hoc tests, statistically significant differences in flexure strength were found in RCBs (BC, GB, and TC) compared to all direct composite resins. Flexural modulus and Vickers microhardness of RCBs (BC, GB, TC) were also significantly different from the direct composite resin (BET, BED, and TEC), except when comparing GS and GB for microhardness. In conclusion, differences between RCBs and direct composite resins were observed regarding flexural strength, flexural modulus, and microhardness, revealing that RCBs have enhanced mechanical properties compared to direct composite resins.

Keywords: CAD/CAM; composite resins; elastic modulus; flexural strength; mechanical properties; microhardness



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1. Introduction

In an era of restorative conservative techniques, the large amount of products available on the market hinders the selection of dental materials [1]. Therefore, particularly when approaching challenging clinical situations, such as the presence of parafunctions or severe dental wear, the most adequate properties and better cost/performance relation should be taken into account.

In the oral environment, restorative materials are subjected to complex masticatory forces. Thus, mechanical properties are one of the main parameters to consider regarding material selection, since they act as an indicator of the restorative material's quality and

behavior [2]. Conventionally, the mechanical properties of ceramic and composite resin materials are evaluated using tests specified by international standards [3]. Flexural strength, elastic modulus, and microhardness are some of the most important properties to consider in restorative materials used to support the occlusal forces of posterior teeth [1,4].

When evaluating the two main groups of dental restorative materials, ceramic restorations have demonstrated higher biocompatibility, color stability, and wear resistance compared to resin-based composites, which are known for their ease of milling and simple intraoral repair techniques [5]. Although direct composite resins are usually effective in addressing clinical scenarios with minor structure loss, in cases of severe loss other options must be considered.

With the introduction of chairside computer-aided design and computer-aided manufacturing (CAD/CAM) technology, new composite resin materials emerged, the resin composite blocks (RCBs), resulting in an optimized alternative option for indirect composite restorations [6]. Industrial processes used to produce CAD-CAM blocks increase material homogeneity and reliability [2,6], decreasing the presence of flaws and pores in comparison to conventional composite resins [7,8].

RCBs are easily milled, exhibit better marginal quality, and can be repaired intraorally, polished, and adjusted for occlusion [9,10]. Additionally, CAD-CAM blocks present an enhanced degree of monomer conversion and have no photoinitiators [11], as they are submitted to industrial polymerization processes [6,11]. Consequently, this material is associated with lower monomer elution, resulting in higher biocompatibility [11].

The wear resistance of restorative materials ensures the stability of occlusal contacts over time and should be similar to natural dentition resistance [2]. RCBs are believed to cause less enamel wear than ceramic restorations [2,12], meaning that the damage to the opposing enamel is minimized [10]. Resin composite blocks have an elasticity modulus similar to dentin, the hardness between enamel and dentin, and high resilience and flexibility to buffer masticatory pressure, allowing them to sustain the most exigent clinical situations [6,9,13].

It is noteworthy that *in vivo* studies show that direct and CAD/CAM composite resin restorations present similar clinical performances [14,15]. Literature that establishes a connection between the *in vitro* results of composite resins and their clinical performance primarily highlights the correlation between fracture toughness and clinical fractures, as well as flexural strength and clinical wear [16]. Therefore, the *in vitro* evaluation of these restorative materials can assess in clarifying whether the RCBs can yield successful clinical outcomes.

Thus, this study aims to compare the mechanical properties (flexural strength, flexural modulus, and Vickers microhardness) of RCBs with direct composite resins, from the same manufacturer. There are two null hypotheses in this study: (H0) There is no difference between the CAD/CAM resin composite blocks and the direct composite resins for the flexural parameters tested. (H1) There is no difference between the CAD/CAM resin composite blocks and the direct composite resins for microhardness.

2. Materials and Methods

2.1. Sample Preparation

Three different resin composite blocks and four composite resins were tested in this study. Experimental groups and information presented in the companies' technical datasheet are presented in Table 1.

Resin composite blocks were sectioned using a precision cutting machine with a diamond disk with 0.3 mm thickness (Accutom 5, Struers, Ballerup, Denmark) with a feed speed of 0.050 mm/s, at 1000 rpm, under permanent water refrigeration. The blocks were firstly sectioned with a parallel cut to their long axis and then rotated 90° to be sectioned perpendicularly (Figure 1a). Therefore, sticks with nominal sizes of 2.1 × 4.2 × 16 mm (depth × width × height) were obtained, as recommended according to ISO 6872:2023 [17].

The outer sticks of each block were discarded, as well as those presenting visible chipping, since these flaws could significantly influence the test results.

Table 1. Evaluated resin composite blocks and composite resins.

Experimental Group		Type of Material	Composition	LOT	Brand
G1	TC	Block: Tetric [®] CAD MT A2/C14	Bis-GMA, Bis-EMA, TEGDMA, UDMA, barium aluminum silicate glass, and silicon dioxide fillers (71 wt. %/51 vol-%)	X51323	Ivoclar Vivadent, Ellwangen, Germany
G2	TE	Composite resin: Tetric EvoCeram [®] A1	Bis-GMA, urethane dimethacrylate, thoxylated Bis-EMA, barium glass filler, ytterbiumtrifluoride, mixed oxide, and prepolymers (75–76 wt. %/53–55 vol-%)	X31979	
G3	GB	Block: Grandio [®] blocs 14L A2 LT	Nanohybrid fillers, UDMA+DMA (86 wt. %/NR)	1821398	VOCO GmbH, Cuxhaven, Germany
G4	GS	Composite resin: Grandio [®] SO A2	Bis-GMA, Bis-EMA, TEGDMA, functionalized silicon dioxide nanoparticles, and glass ceramic filler (89 wt. %/NR)	1847313	
G5	BC	Block: Brilliant Crios [®] CAD/CAM A2 LT 14	Cross-linked methacrylates (Bis-GMA, BIS-EMA, TEGDMA), barium glass and silica particles (71 wt. %/51.5 vol-%)	J27358	Coltene/Whaledent, Langenau, Germany
G6	BET	Composite resin: Brilliant EverGlow [™] Translucent	Bis-GMA, TEGDMA, Bis-EMA, prepolymerized particles containing glass and nano-silica,	180057	
G7	BEU	Composite resin: Brilliant EverGlow [™] Universal Shade A2/B2	aggregated and non-aggregated colloidal silica, and barium glass (71 wt. %/64 vol-%)	I36341	

Abbreviations: NR: not reported; Bis-GMA: bisphenol-A-diglycidyl methacryl; Bis-EMA: ethoxylated bisphenol-Adiglycidyl methacrylate; TEGDMA: triethylenglycol dimethacrylate; UDMA: urethane dimethacrylate; UDMA+DMA: urethane dimethacrylate + dimethacrylate.

On the other hand, direct composite resins were placed in a bar-shaped silicone mold (Aquasil soft putty, Dentsply Sirona, Milford, MA, USA) with nominal sizes of 2.1 × 4.2 × 16 mm (depth × width × height)—(Figure 1b). The specimens were light-cured against a glass plate (Bluephase Style 20i, Ivoclar Vivadent, Lichenstein, 800 mW/cm²) and after demolding, glycerin was applied prior to a second light-curing period of 30 s in a curing chamber (TRIAD 2000, Dentsply Sirona, York, PA, USA), to avoid the oxygen inhibited layer. It was ensured that all specimens were homogeneous and without the presence of voids, clefts, or air blows when viewed radiographically and with magnification. A total of twelve ($n = 12$) samples were obtained per group [17].

All specimens were measured using a digital micrometer with an accuracy of 0.01 mm (Mitutoyo, 156-105, Naucalpan de Juárez, Mexico) and those without-of-range dimensions (± 0.50 mm) were discarded. The tension sides were sequentially polished with 320, 500, and 1200 grit silicon-carbide (SiC) abrasive paper (WSFlex 18-B, Hermes Schleifmittel GmbH, Hamburg, Germany), for 60 s, under running water. Subsequently, specimens were divided into seven groups, respectively identified on the top with a different color mark, and then stored in distilled water, at 37 °C, for 24 h, prior to flexural and microhardness tests.

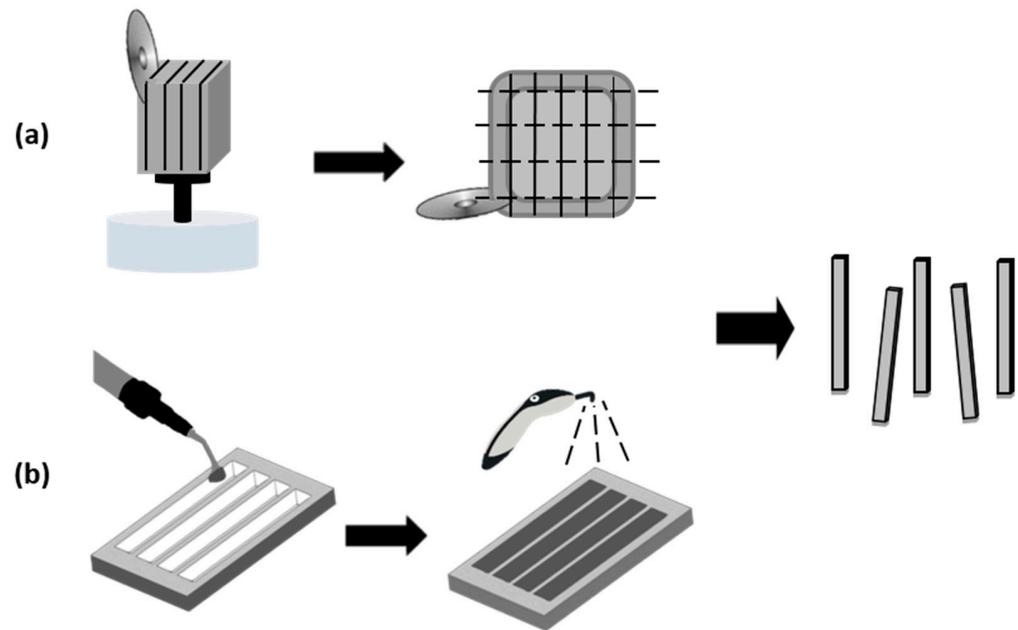


Figure 1. Schematic representation of sample preparation for (a) resin composite blocks and (b) direct composite resin.

2.2. Three-Point Bending Flexural Test

Three-point bending flexural test was performed at a 12 mm span length in an apparatus consisting of three rods (3.0 mm in diameter)—(Figure 2). The specimens were loaded at a constant crosshead speed of 1 ± 0.5 mm/min in a universal testing machine (Model AG-I, Shimadzu Corporation, Kyoto, Japan). The flexure strength (σ) and flexural modulus (E) were calculated, using the software Trapezium (Shimadzu Corporation, Kyoto, Japan), at the maximum flexure/fracture load on the load–displacement curve using the following equations:

$$\sigma = \frac{3FL}{2wt^2} \quad \text{and} \quad E = \frac{FL^3}{4dwt^3},$$

in which F is the maximum flexure load (N), L is the length of the support span (12 mm), w is the width of the specimen (4.2 mm), t is the thickness of the specimen (2.1 mm), and d is the deflection at the load F (mm).

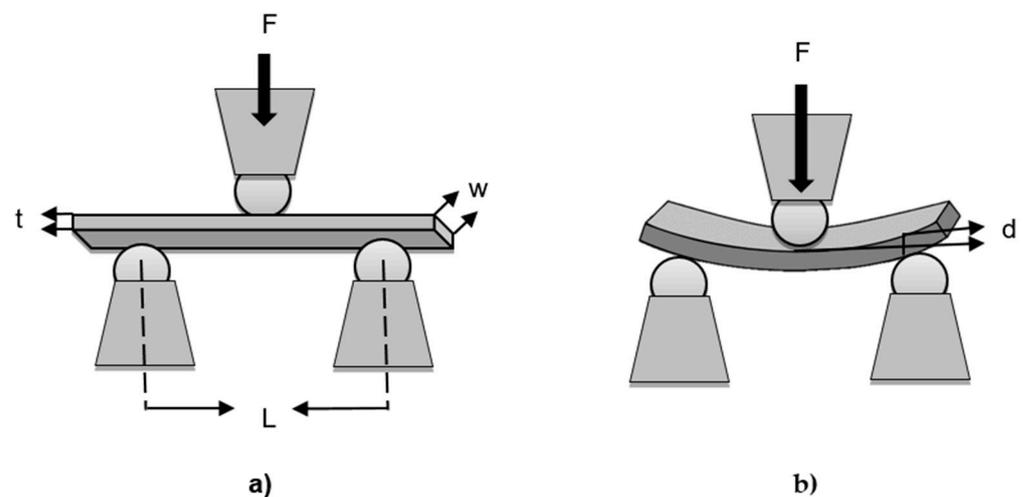


Figure 2. (a) The schematic presentation of three-point bending flexural test execution and (b) the deflection obtained by bending.

2.3. Vickers Microhardness Measurement

After the flexural test, two additional specimens from each group were prepared for the Vickers microhardness test. Five indents were made in each specimen near the center, in a straight line (Figure 3). The distance between the indentations was calculated by multiplying the average indentation diagonal length (D) by four ($4 \times D$) to ensure sufficient distance between them. A microhardness tester (HVM-2, Shimadzu Corporation, Kyoto, Japan) was used with a load of 9.81 N and a dwell time of 15 s based on recommendations of ASTM C1327 [18]. The excessive indentation tips and sides and asymmetric indentations were rejected for measurement.

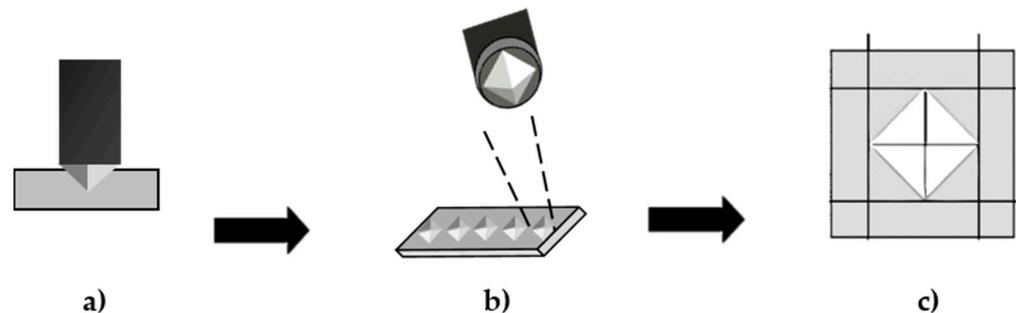


Figure 3. The schematic presentation of Vickers microhardness determination. (a) Indentation made using a Vickers indenter (transversal view); (b) five indentations per sample; (c) measurement of the indentation size (top view).

The major diameters of the Vickers indent (d_1 and d_2) of the square-shaped indentation were measured with light microscopy, and hardness was calculated with the following formula:

$$VHN = (0.102) \times \frac{1.854 F}{D^2},$$

where F is the applied load (kg) and D is the average of the two indentation diagonal lengths (mm).

2.4. Statistical Analysis

Statistical analysis was performed with the IBM SPSS Statistics 23.0[®] program (SPSS Inc., Chicago, IL, USA). Descriptive statistics, such as means and standard deviations, were computed for each property (flexural strength, flexural modulus, and Vickers microhardness) within each material. Normality Shapiro–Wilk (p -value to reject 0.050) and equal variance Levene test (p -value to reject 0.050) were used on the datasets prior to comparison. One-way analysis of variance (ANOVA) was used to compare means of flexural strength followed by the analysis of post hoc pairwise comparisons performed using the Bonferroni correction to determine which specific pairs of means were significantly different. The flexural modulus and Vickers microhardness datasets were analyzed using Welch's ANOVA robust test of equality of means followed by post hoc pairwise comparisons performed using the correction Games–Howell to determine which specific pairs of means were significantly different. Statistical tests were two-sided and were performed using a significance level of 0.05.

3. Results

For flexure strength, one-way ANOVA revealed statistically significant differences amongst groups ($p < 0.01$). Although flexural modulus datasets failed the equal variance Levene test, analysis using Welch's test revealed that all groups had significant equal means ($p < 0.01$). Pairwise comparisons between groups indicated significant differences among all groups except between G1–G5, G2–G6, G2–G7, and G6–G7, both for flexural strength and flexural modulus. RCBs presented higher statistically significant values than the respective direct composite resin, from the same manufacturer, for flexural strength and flexural

modulus. All the tested specimen load–deflection curves were recorded until failure at the maximum load, being observed that all materials underwent a brittle failure. RCB specimens had curves with higher maximum flexure load values than the direct composite resins. The microhardness datasets failed the equal variance Levene test, leading to an analysis with Welch’s test which revealed that all groups had significant equal means ($p < 0.01$). Statistically significant differences among groups were not found between G1 and G5, G2 and G6, G2 and G7, G3 and G4, and G6 and G7. Flexure strength, flexure modulus, and Vickers microhardness values are depicted in Table 2.

Table 2. Flexure strength, flexure modulus, and Vickers microhardness values.

	Flexural Strength (MPa)		Flexural Modulus (GPa)		Vickers Microhardness (HV)	
	mean ± std. deviation	95% Confidence Interval for Mean	mean ± std. deviation	95% Confidence Interval for Mean	mean ± std. deviation	95% Confidence Interval for Mean
		lower bound/upper bound		lower bound/upper bound		lower bound/upper bound
G1	192.4 ± 9.8 ^a	187.0/197.9	15.8 ± 0.3 ^a	15.7/16.0	79.3 ± 6.3 ^a	74.8/83.8
G2	84.1 ± 7.6 ^b	78.6/89.6	12.3 ± 0.9 ^b	11.6/13.0	57.2 ± 7.1 ^b	52.1/62.3
G3	246.5 ± 12.5 ^c	239.6/253.4	20.3 ± 0.7 ^c	19.9/20.7	136.2 ± 9.6 ^c	129.0/143.1
G4	129.6 ± 13.2 ^d	122.3/137.0	18.0 ± 1.3 ^d	17.3/18.7	121.2 ± 11.0 ^c	113.4/129.0
G5	192.8 ± 14.5 ^a	184.8/200.9	15.1 ± 1.5 ^a	14.3/16.0	71.7 ± 3.7 ^a	69.0/72.3
G6	92.5 ± 18.7 ^b	79.2/105.9	11.4 ± 0.8 ^b	10.8/11.9	49.9 ± 7.2 ^b	44.2/54.5
G7	81.1 ± 12.2 ^b	72.3/89.8	10.6 ± 1.4 ^b	9.6/11.6	50.9 ± 14.5 ^b	40.6/61.3
p^*	<0.001		<0.001		<0.001	

* Within columns, similar superscript letters indicate groups that do not present statistically significant differences ($p > 0.05$).

Grandio blocs (G3) and GrandioSO (G4) recorded the highest flexure strength and flexure modulus values for RCBs and direct composite resins, respectively (Figure 4). By ordering the tested groups according to flexure strength, it was found that G3 > G5 > G1 > G4 > G6 > G2 > G7. Additionally, ranking the groups based on flexure modulus leads to the conclusion that G3 > G4 > G1 > G5 > G2 > G6 > G7.

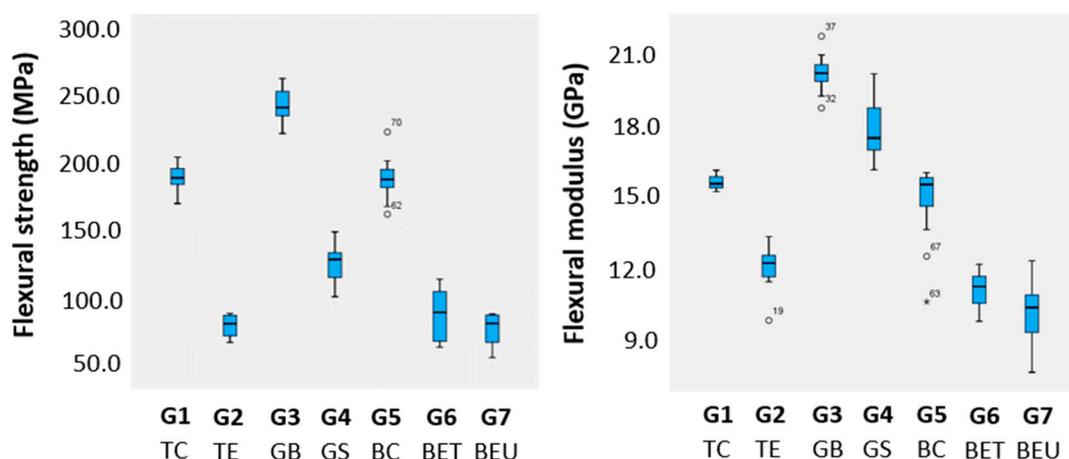


Figure 4. Boxplots of flexural strength and flexural modulus results. Outliers and extreme outliers are represented with dots (o) and asterisks (*), respectively.

Regarding flexure strength and flexure modulus values, the Shapiro–Wilk test detected no departures from normality. The homogeneity of variances was only verified for flexural strength (Levene test, $p > 0.05$).

Similarly to flexural strength and flexural modulus results, Group 3 and Group 4 recorded the highest microhardness values, within each material (Figure 5). Ranking the experimental groups based on microhardness shows that $G3 > G4 > G1 > G5 > G2 > G7 > G6$. Regarding microhardness values, the Shapiro–Wilk test did not detect departures from normality.

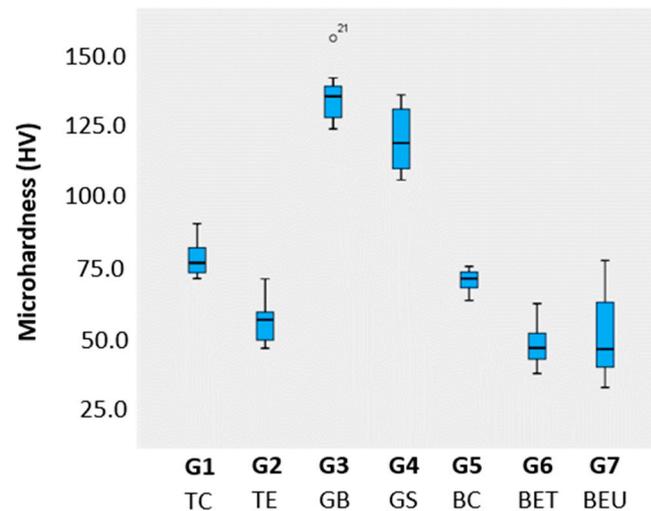


Figure 5. Boxplots of the Vickers microhardness results. Outliers that are represented with dots (o).

4. Discussion

The null hypothesis, (H₀) there is no difference between the CAD/CAM resin composite blocks and the direct composite resins for the flexural parameters tested, and (H₁) there is no difference between the CAD/CAM resin composite blocks and the direct composite resins for microhardness, were both rejected by the findings of this study.

The preparation of resin composite block specimens was conducted according to ISO 6872:2023 [17], a standard procedure usually used to measure the flexural properties of dental ceramics, due to the limitation of CAD/CAM block size. Therefore, it is recommended that a specific test protocol for these materials is developed to precisely measure their flexural properties [19]. Direct composite resin samples were prepared with the same dimensions as resin composite blocks to allow the direct comparison of mechanical properties. All experimental groups had twelve specimens each, taking into account the minimum specified in ISO standards [17].

Over the last few years, extensive research has delved into the numerous correlations between mechanical properties and intraoral performance, allowing a thorough understanding of the importance of materials' mechanical evaluation [20]. Flexural strength determines the maximum stress a restoration is able to withstand prior to failure, when submitted to bending loads [3,4], acting as a great predictor of clinical success and longevity [21]. Additionally, flexural strength has been previously correlated with clinical wear [16]. It should be noted that high flexural strength is desired for restorations that are submitted to large masticatory stress, at least 80 MPa, as required according to the ISO standards for restorative materials [22]. Another important mechanical parameter provided by the flexural test is the modulus of elasticity, which consists of the resistance to elastic deformation under load and defines the rigidity of the composite reconstruction. Composite resin intended for posterior use should have a high modulus of elasticity to maintain the integrity of the adhesive interface, ideally matching or exceeding one of the tissues that it aims to replace, namely dentin (approximately 18.5 GPa).

In the present study, resin composite blocks performed better regarding flexural strength and flexural modulus measurements than direct composite resins. It is noteworthy that although the materials are from the same manufacturer, their composition is different, as displayed in Table 1. From all materials, Grandio blocs (RCB) rendered the highest

values of flexural strength (246.5 MPa) and flexural modulus (20.3 GPa). GrandioSO, from the same manufacturer, presented the highest values of flexural strength from the direct composite resins (129.6 MPa) and the second-highest flexural modulus value of all materials (18.0 GPa). No statistical differences were found between the direct composite resins Brilliant Everglow Trans (BET), used as enamel, and Brilliant Everglow Universal shade (BEU), used as dentin.

In several recent studies with similar methodologies that evaluated different CAD/CAM blocks, RCBs showed lower flexural strength and elastic modulus in comparison to CAD/CAM ceramic and higher values than conventional composite resins, which is in accordance with the results of this study [23–26]. Alamoush et al. evaluated nine different CAD/CAM blocks, concluding that RCBs such as Brilliant Crios (10.98 GPa) and Grandio Blocs (14.8 GPa) have an elastic modulus similar to dentin but lower than enamel's [24]. In another study, Stawarczyk et al. tested eight CAD/CAM blocks and concluded that RCBs such as Lava Ultimate (205 MPa), Cerasmart (184 MPa), and Shofu Block (180 MPa) showed higher flexural strength than leucite ceramic IPS Empress CAD (151 MPa) and the hybrid material VITA Enamic (146 MPa), but lower values than lithium disilicate ceramic IPS e.max CAD (356 MPa) [27].

Besides the three-point bending flexural test, the specimens were also submitted to the Vickers microhardness test, both presenting high reliability. Once more, ten indentations by group were made, as specified in the international standards [18]. Microhardness evaluation is an indicator of the material's wear resistance [3], reflecting the relative ease of finishing and polishing [23]. However, a direct relationship between hardness and wear cannot always be established, due to the multifactorial nature of the wear process [3,23,28]. The main goal is for the material to exhibit enough hardness to resist erosion or/and abrasion by other substances, while simultaneously avoiding wear on the natural tooth surface [29]. Therefore, the restorative material's hardness should match that of enamel [30].

Several studies investigated the hardness of different materials showing lower Vickers hardness values for the composite resin nanoceramics when compared to hybrid and ceramic blocks [23,24,27,28,31]. In the present study, Grandio blocs (136.2 HV) and GrandioSO (122.1 HV) had the highest microhardness values and Brilliant Everglow Translucent and Universal shade (49.9 and 50.9 HV, respectively) had the lowest values. With the exception of GrandioSO, resin composite blocks rendered higher values than direct composite resins.

The reported superior mechanical properties of CAD/CAM composite resin can be attributed to the fact that the higher degree of conversion is associated with an increase in wear resistance, hardness, and elasticity modulus [7]. In addition, it is noteworthy that resin composite blocks present smaller interparticle space in comparison to direct composite resins [2], leading to a more uniform stress distribution [32]. Despite the variations in the morphology observed in SEM analyses, there are no statistically significant differences regarding biofilm formation between direct composite resins and RCBs, from the same manufacturer [33].

In addition to the polymerization process, the filler content also influences the mechanical behavior of restorative materials [7,24,31,32,34]. Amongst the studied materials, Grandio Blocs and GradioSO present the highest filler content, which resulted in enhanced mechanical properties, in each respective category. In accordance with the present results, Papathanasiou et al. concluded that Grandio Blocs were associated with the highest elastic modulus and hardness values, due to the superior filler content [8]. Furthermore, Alamoush et al. reported that the resin-matrix composition directly influences the degree of conversion and the water sorption, with Grandio Blocs rendering the lowest water sorption values among the tested materials [35].

The use of spherical-shaped filler particles is also associated with increased volume fraction due to the particles' higher compaction. The addition to the resin matrix of quartz, barium glass, and silica, which are present in all the composite resins used in this study, enhances wear resistance by reducing the particle size and increasing the filler volume [36].

From a biomimetic point of view, composite resins hold considerable value and are extensively used in daily clinical practice. However, the rehabilitation of structurally compromised teeth or extensive oral rehabilitations requiring the stability of the vertical dimension of occlusion often requires restorative materials with superior mechanical properties [23,25,27,31]. Therefore, resin composite blocks are an alternative, as this material closely mimics dental tissue properties and is able to absorb functional loads [24,27,31]. Their clinical application encompasses single-tooth restorations, both in the anterior and posterior regions, such as inlays, onlays, crowns, and veneers. Nevertheless, the process of scanning and milling indirect restorations is more time-consuming compared to direct restorations, cured intra-orally. The cost-benefit analysis for both alternatives is therefore debatable and contingent on the specific requirements of each particular clinical case [37].

Overall, whenever there is an indication for an indirect restoration in composite resin, this restoration should be made with resin composite blocks. However, regarding small/medium restorations of vital teeth, the literature seems to indicate that the performance of direct composite resin restorations is effective [38]. Additionally to the amount of tooth structure to be restored, the adhesion values between the dental tissue and the CAD/CAM blocks should also be further investigated.

The observed significant differences found between the evaluated materials can be due to the intrinsic limitations of the manufacturing process or to the conditions inherent to the fabrication process of the specimens with both materials. Future in vitro and clinical studies are necessary to gain a more comprehensive understanding of these material characteristics and unveil their clinical potential and performance over time. Additionally, different properties and various composite resins with distinct organic and structural compositions should be investigated.

5. Conclusions

Within the limitations of this in vitro study, the following conclusions were drawn:

- The mean flexural strength of the RCBs (BC, GB, and TC) was significantly higher than all direct composite resins tested (BET, BEU, GS, and TEC).
- The mean flexural modulus of the RCBs (BC, GB, TC) and the direct composite resin GS was significantly higher than the other direct composite resin tested (BET, BED, and TEC).
- The mean microhardness of the RCBs (BC, GB, TC) and the direct resin GS was significantly higher than the other direct composite resin tested (BET, BED, and TEC).
- The RCB and the direct resin composite evaluated in this study with better mechanical properties were Grandio blocs and GrandioSO, respectively.

Author Contributions: Conceptualization, J.C.R. and R.D.; methodology, J.C.R. and A.M. (Ana Messias); software, A.M. (Alfredo Marinho) and A.M. (Ana Messias); validation, J.C.R., R.D. and A.M. (Alfredo Marinho); formal analysis, A.V. and G.A.; investigation, J.C.R. and A.M. (Alfredo Marinho); resources, A.M. (Ana Messias); data curation, G.A. and A.M. (Ana Messias); writing—original draft preparation, A.M. (Alfredo Marinho); writing—review and editing, G.A.; visualization, R.D.; supervision, A.V. and G.A.; project administration, J.C.R. and R.D. All authors have read and agreed to the published version of the manuscript.

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