



Article

Colour Changes of Acetal Resins (CAD-CAM) In Vivo

Cristina Gómez-Polo ^{1,*}, Ana María Martín Casado ², Norberto Quispe ¹ , Eva Rosel Gallardo ³
and Javier Montero ¹ 

¹ Department of Surgery, Faculty of Medicine, University of Salamanca, 37007 Salamanca, Spain

² Department of Statistics, Faculty of Medicine, University of Salamanca, 37007 Salamanca, Spain

³ Department of Preventive and Community Dentistry, Faculty of Dentistry, University of Granada, 18011 Granada, Spain

* Correspondence: crisgodent@usal.es; Tel.: +34-923294500 (ext. 1996)

Abstract: To quantify the discolouration of the temporary acetal resins in vivo, based on the weeks of follow-up and the salivary pH in the three thirds of the tooth. To find out if the final CIELAB coordinates can be predicted from the initial colour coordinates, the salivary pH, the situation (in thirds) and the weeks of follow-up. Colour coordinates (L, C, and h) were recorded by spectrophotometry in 13 participants fitted with hybrid provisional complete dentures made of acetal resin. Colour recordings were made on the day of placement and after several weeks of follow-up (6 to 31 weeks). Salivary pH was also measured as a predictor variable for colour change. The ANOVA statistical test and regression models have been used. The highest colour difference according to ΔE_{ab}^* was 27.46 units after 15 weeks of follow-up and the lowest was 7.34 units after 17 weeks of follow-up. Neither in the cervical nor in the middle third any regressor variable (initial L*, initial C*, initial h*, salivary pH and weeks of follow-up) was able to significantly predict any of the final colour coordinates ($p > 0.05$). The colour change of the temporary acetal resins used exceeds the threshold of clinical acceptability, and it is not acceptable to maintain satisfactory aesthetics. The weeks of follow-up and the salivary pH are not capable of satisfactorily predicting the final color coordinates of the acetal resins.

Keywords: acetal resins; spectrophotometer; CIELAB colour space; difference of colour formulae; interim dental restoration; dentistry; temporal resin



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1. Introduction

Tooth colour affects the attractiveness of an individual's smile; if it is not pleasing, it can lead to rejection and discomfort on the part of the patient and their environment. People with bright teeth are associated with better personal, social and economic characteristics [1]. One of the factors influencing the perception of smile attractiveness is gender and dental symmetry [2]. Although the vast majority of research indicates that the most valued aesthetic factor is tooth colour [1,3,4]. The materials used to fabricate temporary prosthetic restorations are becoming increasingly important in the treatment plan, mainly because they are made more quickly and allow the patient to assess the shape, colour, position, and size of the teeth before the final dental prosthesis is made. All provisional materials must maintain acceptable aesthetics and resemble the natural tooth for an adequate period of time [5]. The success of dental prosthetic restorations includes patient satisfaction with aesthetics. The constant evolution of dental restorative materials has increased the selection options for the clinician. It is possible to use everything from classical metal alloys and ceramics, through zirconia, leucite, lithium disilicate, which have appeared more recently as definitive materials, to temporary materials derived from acrylic resins or hybrids [6]. Temporary dental prosthesis is defined as "a fixed or removable dental prosthesis, or maxillofacial prosthesis designed to enhance aesthetics, stabilization, and/or function for a limited period of time, after which it is to be replaced by a definitive dental

or maxillofacial prosthesis; often such prostheses are used to assist in determination of the therapeutic effectiveness of a specific treatment plan or the form and function of the planned definitive prosthesis" [7]. The proper fabrication of a temporary fixed prosthesis plays a major role in the success or failure of definitive restorative treatment. Implant-supported fixed provisional dental restorations, especially in fully edentulous patients, serve as a mock-up to assess whether the more costly definitive fixed dental restorations fit the patient's aesthetic needs and preferences, providing a predictability factor. On the one hand, the patient "tries in" the provisional fixed prosthesis and can express their satisfaction or dissatisfaction. On the other hand, the clinician assesses the adaptability of the dental and/or gingival structures [8] and thus generates a more approximate judgement to modify the treatment plan if necessary, minimising costs. Nowadays, the high aesthetic demands and the reduction of waiting times to get patients' teeth in place have led to a remarkable improvement in the field of provisional restorations. Temporary restorations can be fabricated using conventional techniques (often self-curing polymeric materials) and by CAD/CAM (Computer-aided design/Computer-aided manufacturing) techniques [9,10]. The materials used for the fabrication of provisional prostheses need to fulfil certain characteristics: they must preserve the health of adjacent periodontal tissues, be biologically inert, resist masticatory functional loads, offer adjusted handling and working time, provide good occlusion, adequate phonetics, satisfactory aesthetics, colour stability, low cost, speed of fabrication, and ideally, offer the possibility of being repaired/modified [11,12]. Within the group of temporary materials derived from acrylic resins, one of the most widely used and studied is polymethylmethacrylate (PMMA) [9,13]. Acetal resins have recently been introduced for the purpose of making temporary dental prostheses by CAD-CAM, although their use is not yet widespread. Acetal resins are thermoplastic polymers derived from formaldehyde. They are usually coloured to more closely mimic tooth colour, as their natural colour is white. The homopolymer of acetal resin is called polyoxymethylene (POM) and its composition is a chain of methyl groups alternating with oxygen molecules [14]. In the surgical-medical field, acetal resins have already been frequently used to make hip prostheses and heart valves. In dentistry, they appeared as an alternative to the poor aesthetics of metal retainers in removable acrylic partial dentures [14,15] and as an alternative material for patients who are allergic to any of the components of chrome-cobalt alloys [16]. POM has favourable properties [14], such as resistance to deformation [16], wear [17] and water absorption [18]. Few studies have been published on the quality of POM as a temporary restorative material and on assessing its colour stability [9,11]. Depending on the results obtained on its colour behaviour, it might be possible to recommend its use for a longer period of time.

To quantify tooth colour, colourimeters and spectrophotometers [19,20] are used to provide colour results in coordinates in the CIELAB color space [21] system. The CIELAB color space consists of 3 coordinates: (1) L^* (lightness) measures the amount of black and white, (2) a^* coordinate (green-red axis) and (3) b^* coordinate (blue-yellow axis)*. They are interpreted as follows: the higher the L^* coordinate, the greater the amount of white, which gives a brighter tooth; the higher the a^* coordinate, the greater the amount of red; the higher the numerical value of the b^* coordinate, the greater the amount of yellow [22]. Subsequently, with the aim of each coordinate to represent one of the three dimensions of colour (Lightness, Chroma, and hue), the CIELCh color space appeared. In this nomenclature, the L^* coordinate remains identical, representing the lightness*, the C^* coordinate represents the chroma (amount of hue) and the angular coordinate h^* represents the hue (circular axis; the units were in the form of degrees (or angles), ranging from 0° (red) through 90° (yellow), 180° (green), 270° (blue) [22]. In the dental field, two formulas have been used to calculate the difference between two colours: (1) Classical Euclidean Formula widely used in dentistry whose equation is $\Delta E_{ab}^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$. ΔE_{ab}^* represented the magnitude of the colour difference [23] and (2) the CIEDE2000 formula

($\Delta E00^*$) [23,24] which is closer to the threshold for colour discrimination than the formula ΔEab^* and is better suited for interpreting clinical results [25].

$$\Delta E00 = \left[\left(\frac{\Delta L'}{K_L S_L} \right)^2 + \left(\frac{\Delta C'}{K_C S_C} \right)^2 + \left(\frac{\Delta H'}{K_H S_H} \right)^2 + R_T \left(\frac{\Delta C'}{K_C S_C} \right) \left(\frac{\Delta H'}{K_H S_H} \right) \right]^{1/2}$$

Several research studies have focused on the study of colour in the dental environment through the perceptibility and acceptability threshold (colour differences that can be perceived and accepted by 50% of the observers) [26,27], in order to categorise colour changes. Several factors are involved in the discolouration of temporary dental restorative materials, including: the composition of saliva (immunoglobulins, enzymes, nitrogenous products and mucins), salivary pH [28] ranging from 5.8 to 7.1 (slightly acidic) [29], and exogenous colouring substances [30,31].

The aim of the present research in vivo was:

1. To study whether the degree of discolouration or difference of color, of acetal resin (POM) used as a provisional material in complete upper hybrid prostheses, is significantly related to the weeks of follow-up and to salivary pH in the three thirds of the tooth (gingival third (area 1), middle third (area 2), and incisal third (area 3)).
2. To find out if the final CIELAB coordinates can be predicted from the initial colour coordinates, the salivary pH, the situation (in thirds) and the weeks of follow-up.

The null hypothesis stated was (1) that there is no colour change ($\Delta Eab^*/\Delta E00$) in the implant-supported fixed provisional restorations made of acetal resins (POM) below the dental chromatic acceptance limit and (2) the final CIELAB coordinates can be predicted in thirds, from the initial colour coordinates, the salivary pH and the weeks of follow-up.

2. Materials and Methods

A total of 13 subjects were included in the study to undergo complete rehabilitation with temporary fixed tooth-supported hybrid prostheses with acetal resins in the upper jaw. All hybrid provisional complete prosthesis made of acetal resin were designed with facebow and a semi-adjustable articulator. Inclusion criteria for this study were: being an adult, of full mental capacity, in good general health and with availability of time to attend check-ups. Subjects with severe bruxism and those with diagnosed gastric reflux were excluded. All patients signed the informed consent form. This research was conducted in accordance with the ethical principles of the World Medical Association Declaration of Helsinki. The experimental protocol was approved by the Bioethics Committee of the University (201500006834). Polyoxymethylene (POM, Definifit, GT Medical, Spain) was used as a temporary restorative material; POM is a resin intended for the fabrication of temporary dental prostheses, according to its manufacturer. The POM discs used in this study were machined with fine-grained tungsten carbide burs, following the manufacturer's instructions. For the fabrication of the POM hybrid prostheses, the concept proposed by Kapos et al. [32] of "Complete CAD/CAM Product" was used, which means that the final restoration has undergone a fully computerised design and fabrication process throughout; therefore, the prostheses used were fabricated from a pre-polymerised block of POM in its entirety, without interphases. All milled POM discs (CAM) had the same initial colour: A3, according to the Vita Classical guide. After completing the milling process, finishing, and polishing treatments were carried out on the provisional hybrid prostheses, following the manufacturer's instructions: non-aggressive pastes, (Acrypol, Bredent, Senden, Germany), without generating excessive heat and with silicone polishers. The milling and finishing process of all the prostheses was carried out in the same prosthetic laboratory with the same machinery (DWX-4, DCSHAPE, Barcelona, Spain) and by the same laboratory technician with 12 years of experience. Colour coordinates (L^* , C^* , h^* , a^* , and b^*) were recorded by spectrophotometry (Spectroshade Micro, MHT Optic Research, Switzerland) in each of the three thirds of the upper right central incisor: cervical, middle, and incisal edge. Each third of the tooth was measured three times and arithmetic means were used for further statistical

analysis. The tooth colour of the temporary hybrid restoration was measured at two time points: on the day of placement and after several weeks in the mouth (from 6 weeks to a maximum of 31 weeks) (Figure 1). In addition, Euclidian equation and CIEDE200 colour difference formulae were used to calculate the colour difference. In the present study, the reference values for interpreting the colour difference were those published by Douglas et al. with $\Delta E_{ab}^* = 2.6$ units as the threshold of perception and $\Delta E_{ab}^* = 5.5$ units as the threshold of clinical acceptability [27]. The software used for the descriptive and inferential statistical analysis of the results was IBM Corp. Released 2017. IBM SPSS Statistics for Windows, Version 25.0. Armonk, NY: IBM Corp. ANOVA test and linear regression analyses with R squared for indicate how well data fit a statistical model were used. Based on the sample size of similar studies [10,32,33] an initial sample size of 30 subjects was established for detecting clinical discolorations ($\Delta E_{ab}^* > 5.5$ units) in the fixed provisional restorations made of acetal resins [26], but consecutive sampling was discontinued due to the severity of the colour discolorations.

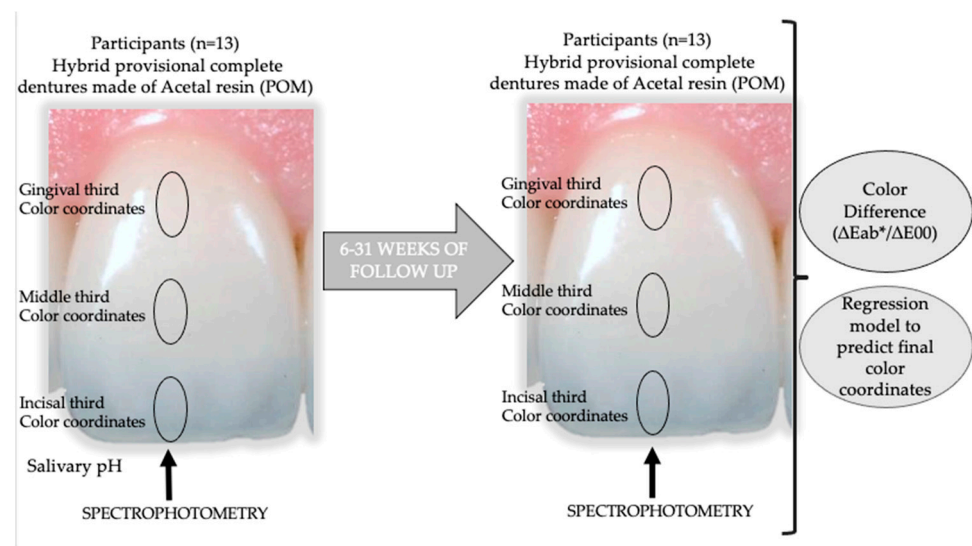


Figure 1. Scheme of the methodology.

3. Results

Table 1 shows the colour changes, according to the ΔE_{ab}^* formula and the ΔE_{00}^* formula, between the day of placement of the provisional hybrid POM prosthesis and after the different weeks of follow-up in the three thirds of the tooth. The highest colour difference according to the ΔE_{ab}^* formula was 27.46 units after 15 weeks of follow-up and occurred in the cervical third. On the other hand, the lowest shade difference was 7.34 units after 17 weeks of follow-up and took place in the incisal third. These colour differences exceeded the threshold of clinical acceptability (Figure 2) proposed by Douglas et al. [27] According to the ΔE_{00}^* formula, the lowest colour change was 5.23 units, and the highest colour change was 22.61 units (Figure 3). The pH range varied from 6 to 9 units and the follow-up weeks ranged from a minimum of 6. to a maximum of 31.

Table 1. Colour differences, according to the Euclidean and CIEDE2000 formulae, in the three thirds of the upper central incisor.

| Participants | CIELAB (ΔE_{ab}^*) | | | CIEDE2000 (ΔE_{00}^*) | | | pH | Weeks of Follow-Up |
|--------------|------------------------------|--------------------------|---------------------------|---------------------------------|--------------------------|---------------------------|-----|--------------------|
| | Cervical ΔE_{ab}^* | Middle ΔE_{ab}^* | Incisal ΔE_{ab}^* | Cervical ΔE_{00}^* | Middle ΔE_{00}^* | Incisal ΔE_{00}^* | | |
| 1 | 24.92 | 25.09 | 19.67 | 21.06 | 20.37 | 16.29 | 6 | 16 |
| 2 | 17.44 | 13.88 | 18.98 | 15.04 | 11.07 | 16.21 | 6.5 | 12 |
| 3 | 18.71 | 19.36 | 20.16 | 15.88 | 16.28 | 17.48 | 7 | 31 |
| 4 | 18.24 | 16.29 | 9.63 | 16.41 | 13.57 | 8.40 | 6 | 14 |
| 5 | 16.01 | 15.13 | 11.10 | 13.75 | 12.23 | 8.81 | 8 | 27 |
| 6 | 15.49 | 17.02 | 14.15 | 13.01 | 13.13 | 10.84 | 9 | 18 |
| 7 | 19.51 | 21.14 | 17.77 | 18.20 | 17.76 | 14.51 | 8.5 | 18 |
| 8 | 11.01 | 12.01 | 11.42 | 8.68 | 9.53 | 9.36 | 6.5 | 6 |
| 9 | 9.87 | 9.21 | 7.34 | 8.16 | 6.72 | 5.23 | 6 | 17 |
| 10 | 18.03 | 16.50 | 13.97 | 14.97 | 13.47 | 11.45 | 9 | 15 |
| 11 | 23.71 | 25.59 | 19.81 | 18.70 | 19.28 | 13.43 | 7.5 | 15 |
| 12 | 10.58 | 10.75 | 9.54 | 7.65 | 7.81 | 6.96 | 8 | 15 |
| 13 | 27.46 | 23.86 | 15.35 | 22.61 | 18.38 | 11.20 | 7.5 | 15 |



Figure 2. Color difference between A3 dental shade tab and hybrid provisional complete prosthesis made of acetal resin.

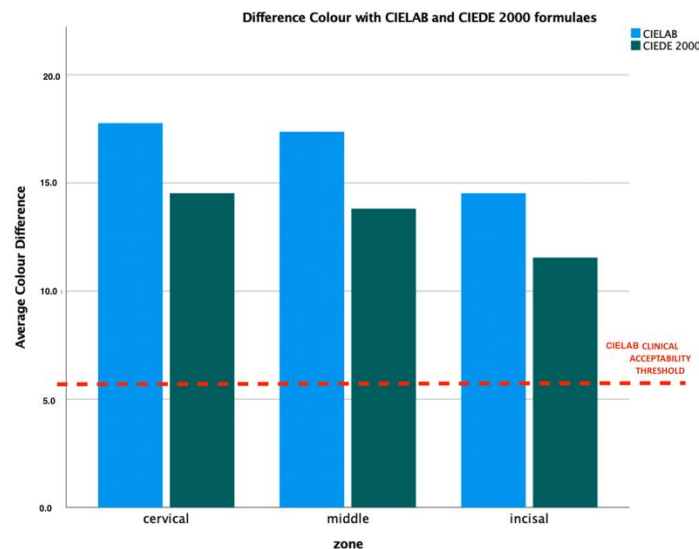


Figure 3. Colour differences (ΔE_{ab}^* and ΔE_{00}^*) exceeded the threshold of clinical acceptability.

With both formulas, the ANOVA with one factor of variation (thirds of the tooth) in a randomised block design (subjects) revealed that the chromatic changes in the three areas differed significantly ($p = 0.002$). Subsequently, Duncan’s test showed that discolourations

in the cervical and middle areas did not differ significantly, but discolourations in the incisal area did, where colour changes were significantly smaller.

By using regression models, it was assessed whether the degree of discolouration was significantly related to the weeks of follow-up and to pH, according to the two colour formulae in each of the tooth thirds.

Euclidean formula (ΔE_{ab}^*):

$$\widehat{Dif}_{cervical} = 16.379 - 0.034pH + 0.097 \text{ Weeks of follow up } (R^2 = 0.012)$$

$$\widehat{Dif}_{middle} = 12.561 + 0.363pH + 0.127 \text{ Weeks of follow up } (R^2 = 0.032)$$

$$\widehat{Dif}_{incisal} = 11.493 + 0.090pH + 0.141 \text{ Weeks of follow up } (R^2 = 0.041)$$

In all three areas, the coefficients of determination were small, which indicates that discolouration depends on explanatory variables other than pH and follow-up weeks. However, an increase in the coefficient of determination was observed from the cervical to the incisal area. No model was significant (tests for significance of regression with $p > 0.05$). It is noteworthy that the signs of the regression coefficients for 'pH' varied according to the area (negative in the cervical area, positive in the middle and incisal area), while the signs of the regression coefficients for the variable 'weeks of follow-up' were the same (positive) in all three areas.

CIEDE2000 formula (ΔE_{00}^*):

$$\widehat{Dif}_{cervical} = 13.729 - 0.099pH + 0.115 \text{ Weeks of follow up } (R^2 = 0.022)$$

$$\widehat{Dif}_{middle} = 9.938 + 0.206pH + 0.140 \text{ Weeks of follow up } (R^2 = 0.047)$$

$$\widehat{Dif}_{incisal} = 10.467 - 0.211pH + 0.157 \text{ Weeks of follow up } (R^2 = 0.062)$$

In all three zones, the coefficients of determination were small (although larger than those of the models constructed with the Euclidean formula), which indicates that discolouration depends on more explanatory variables (apart from salivary pH and weeks of follow-up). Again, an increase in the coefficient of determination was observed from the cervical to the incisal area. No model was significant (tests for significance of regression at $p > 0.05$). The signs of the regression coefficients for 'pH' varied according to the area (negative in the cervical and incisal areas), while the signs of the regression coefficients for the variable 'Weeks of follow-up' were the same (positive) in all three areas.

It was studied whether the final colour coordinates, in each of the three thirds, could be predicted from the initial colour coordinates, pH, salivary and weeks of follow-up. In the cervical and middle third, no regressor variable (initial L^* , initial C^* , initial h^* , salivary pH and follow-up weeks) was able to significantly predict any of the three final colour coordinates: Final L^* , final C^* , and final h^* ($p > 0.05$). In the middle third, no regressor variable (initial L^* , initial C^* , initial h^* , salivary pH and weeks of follow-up) was able to significantly explain the dependent variables ($p > 0.05$). On the contrary, in the incisal third, the independent variable initial C^* had a predictive power of the final C^* coordinate and the model was significant ($p = 0.017$). Similarly, in the incisal third, the independent variable initial h^* had significant explanatory power over the dependent variable final h^* ($p = 0.006$). Thus, in the incisal third, the final intensity (final C^* coordinate) and the final shade (final h^* coordinate) can be explained by the corresponding initial colour coordinates of the acetal resins.

4. Discussion

The first null hypothesis was rejected because all the color differences were above the threshold of dental clinical acceptability ($\Delta E_{ab}^* > 5.5$ units). The null's second hypothesis cannot be accepted either, since the predictive power of the initial color coordinates, salivary pH, and weeks of follow-up have small coefficients of determination.

Many works have studied provisional restorative materials in relation to marginal fit, hardness, roughness, fracture, and strength [9,10,34,35], but few have focused on acetal resins (POM). CAD-CAM technology has also made it possible to achieve an extraordinary improvement in quality based on mechanical strength [36,37], flexural strength [38], better marginal fit [39], fewer imperfections [40] and improved aesthetics [41,42]. There are contradictory results such as those published by Tieh et al. who claim that the colour stability of CAD-CAM in denture teeth is similar to that of traditional PMMA denture teeth [43]. In the same line, Stawarczyk et al. concluded, in *in vitro* studies, that resins manufactured by CAD-CAM display the same colour stability as glass-ceramic materials [40]. These differences could be due to the variation in the methodology of the studies and the diversity of Bis-acrylic resins on the market in terms of composition.

According to the manufacturer, Definitfit[®] acetal resins are sensitive to salivary pH below 4, and also if the material is heated excessively during the milling phase, if it is left in contact with carotenes for a long time and if polishing is not carried out properly. In none of the participants was the pH below 6. If the results obtained in *in vivo* studies on shade stability are favourable, acetal resins could be used as a material for long-term temporaries (more than 6 months) [44]. The main aesthetic disadvantage of interim restoration materials is that they undergo absorption and adsorption processes of liquids present in the oral environment, which can cause discolouration and reduce the degree of aesthetic satisfaction of the patient [5,44–46]. *In vitro* studies have concluded that factors such as type of diet, intake of certain medications and mouthwashes influence the degree of staining of different types of polymeric materials [40,47–49]; this is particularly true with the consumption of coffee, tea, and wine [5], which are the beverages most capable of creating discolourations [50] and the intensity of its action is difficult to quantify as it depends on its quality [5,17,51]. This could have explained better the color change of patient #9 that, although higher than clinical acceptable thresholds, showed a marked lower color-change in respect to other patients.

The main disadvantages of the present research may be that the diet and deleterious habits of the participants were not recorded, and these variables could also be responsible for the significant colour changes observed; on the other hand, salivary pH and weeks of follow-up have not been shown to be a determining factor in the colour changes. For this reason, future research would need to broaden the baseline variables (diet and habits) in order to quantify the independent effect on the intraoral discolouration of these materials. In this regard, due to the inherent difficulty of fully controlling the oral diet *in vivo* there are still no published studies that quantify the effect of each food on tooth color. Most of the research had studies beverages (staining solutions) [30]. Also, it is necessary to take into account the different chromatic behavior of food and drinks *in vivo* than *in vitro* [30]. One of the disadvantages of *in vivo* studies is the difficulty of standardising intraoral conditions, because they depend on many factors; however, they bring us closer to the intraoral reality. A small number of *in vivo* studies of dental restorations made of polymeric materials [44,52–54] are available, most of them in the field of denture prosthetics. Despite the small sample size, the primary outcome (change discoloration) was found to be statistically significant between baseline and postoperative evaluations (Table 1), demonstrating that this sample size was enough potent to detect severe change discolorations.

The great variety of materials of this nature on the market as well as the wide range of fabrication techniques, the different methodologies, and the lack of standardisation in evaluation criteria and follow-up times make it difficult to compare their results. The *in vivo* study by Díez-Quijano et al. on implant-supported fixed temporary prosthesis with CAD-CAM acetal resin in posterior sectors, using a colourimeter for colour coordinate registration, revealed that at six months the mean colour change according to the ΔE_{ab}^* formula was 12.90 units. They also found that the h^* -coordinate (hue) and a^* -coordinate decreased significantly at six months compared with baseline ($p < 0.05$). They concluded that PMMA showed better colour behaviour than POM. [11] In the present work, colour was recorded spectrophotometrically on the surface of the upper incisors (which is less

convex than the surface of the posterior sectors). The smallest colour change according to the ΔE_{ab}^* formula was 7.34 units (after 15 weeks of follow-up) and the largest was 27.46 units (after 17 weeks of follow-up). These differences in results may be due to the composition of acetal resins, smoking habits, diet, and different electronic colour-recording devices. Spectrophotometers are known to record the amount of light reflected by an object, but they have the disadvantage of scattering, especially on convex surfaces. In these in vivo studies [15], the average colour change also exceeds the colour acceptability threshold (ΔE_{ab}^* 5.5 units), which prevents the desired colour stability requirement from being met. Most authors publish results according to the Euclidean formula, although the CIEDE2000 formula is more in line with human visual perception [22,55]. In reference to the in vitro colour changes of POM resins not processed by CAD-CAM, we found ΔE^* values lower than 5.4 units [9] after six weeks of immersion in different solutions. Another in vitro study along the same lines by Ozkan et al. [19] compared the colour stability of acetal resin and conventional PMMA subjected to a thermocycling process. Although significant differences in colour change were found at the initial and final time, both materials obtained clinically acceptable ΔE^* values (1.33 units for PMMA and 1.13 units for acetal resin). Caution should be exercised when analysing these colour differences, as the composition of the acetal resin varies, as well as the manufacturing and processing process. It was not possible to predict the colour change that the POM resins will show, depending on the variables analysed, with statistical determination in this research. Only in the incisal third, the dependent variables final C^* and final h^* were found to be significant regressors of initial C^* and initial h^* , respectively. Another aspect to take into account is hygiene habits, which could also play an important role in the maintenance of the initial shade of the POM restoration [14]. Also, Alkhatib et al. [56] found that there is a significant difference between heavy smokers and non-smokers with regard to the presence of discolourations. With reference to saliva, there is an increasing need to study its involvement in discolourations and the influence of salivary enzymes on the degradation of temporary resin materials. [31] Additionally, more studies are needed in order to test also flexural [57] and hardness [58] test, as these variables have a significant influence on material choice. The results presented do not encourage the use of POM resins as a long-term temporary restorative material, as the colour changes observed exceeded the colour acceptance threshold, and this may lead to patient dissatisfaction.

5. Conclusions

With the limitations of this study, we can affirm that the colour change of the acetal resins used is not acceptable to maintain satisfactory aesthetics (exceeds the threshold of clinical acceptability). The discolouration of acetal resins is not only dependent on pH and weeks of follow-up.

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Informed Consent Statement: All participants were volunteers.

Data Availability Statement: Not applicable.

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Conflicts of Interest: The authors declare no conflict of interest.

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