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Journal of Building Material Science
http://ojs.bilpublishing.com/index.php/jbms

ARTICLE

Steatite/Epoxy Composites for Restoration Works Through a Statistical Mixture Design Methodology

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ARTICLE INFO

Article history
Received: 18th February 2019
Accepted: 29th April 2019
Published: 29th April 2019

Keywords:
Planning of experiments
Composites
Restoration
Steatite

ABSTRACT

Currently many works of art made of soapstone and recognised as cultural patrimony of humanity are in an advanced stage of degradation. Hence, it is necessary to interrupt this process and recover the deteriorated parts. Composite materials consisted of steatite particles and epoxy polymer are designed and characterised for their application in the repair of sculptures made of soapstone. The material applied in restorations should provide colouration and texture similar to soapstone besides structural requirements. The degree of similarity of the artificial material to the rock is enhanced by the proper selection of the particle size range and the increase of the weight percent of steatite incorporated in the composites. A statistical methodology based on the mixture design is used to optimise the relative amount of three size of steatite particles in order to maximise the weight percent of dispersed phase in the composites. The maximum particle packing density (1.50 g/cm³) is obtained for a ternary mixture, composed of 62 wt.% of coarse particles (1.18 mm - 0.60 mm), 6 wt.% of medium sized particles (0.60 mm - 0.30 mm) and 32 wt.% of fine particles (0.30 mm - 0.15 mm). In this manner, the fabrication of composites based on an epoxy polymer matrix with 70 wt.% of incorporated steatite particles has been possible, increasing the maximum amount by 10 % as used in previous works.

1. Introduction

The technique of rock carving has been used in the constructions of Brazil since the 16th century, reaching its apex and primarily in the state of Minas Gerais during the 18th century. In this region, the art was implanted by influence of the Portuguese and acquired local peculiarities thanks to the creativity of the native artists. These artists dominated the eighteenth-century architecture and helped to compose the beautiful and original collection that characterises the baroque style of Minas Gerais[1]. During the eighteenth century many cities in the Brazilian´s country side were founded because of the discovery of large gold deposits. The hope for prosperity and enrichment brought by the precious metal attracted large numbers of migrants and resulted in a strong economic development of these regions. Popula-

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 DOI: https://doi.org/10.30564/jcr.v1i1.152

tion growth and abundant financial resources boosted the local economy. These factors, allied to the strong religious tradition of that time in Brazil, resulted in the construction of magnificent churches with ornate facades and countless works of art[2].

In this prosperous scenario, great artists of the baroque appeared; Antonio Francisco Lisboa, popularly known as Aleijadinho, being the most famous. Like Aleijadinho, many sculptors of that time used blocks of rock as the main raw material for the manufacture of sculptures and ornaments of churches. Steatite, also known as soapstone, was the favourite rock material used by these sculptors, because of its low hardness, and, therefore, the great ease of obtaining precise and delicate details of the artefacts. However, this striking feature also turns soapstone susceptible to wear and, unfortunately, to acts of vandalism. Many artefacts made of soapstone, considered as cultural patrimony of humanity, are currently in an advanced stage of degradation[3]. Therefore, it is necessary to develop techniques and actions for the maintenance and restoration of these artefacts. In view of this need, this work aimed to develop suitable composite materials for application in the repair of superficial wear, replacement of damaged parts and production of replicas of sculptures in soapstone.

In this way, studies have been conducted on developing suitable materials to be used in the restoration of artefacts made from soapstone. The material used in such restorations must present, as one of their main characteristic, colouration and texture similar to the original stone, and acceptable structural properties. Therefore, composites made with different weight percent of particles of soapstone itself and different weight percent of binders have been studied in order to obtain a material suitable for restoration works[4][5][6].

Strecker et al.[4] have investigated the effect of adding steatite residues in different grain sizes on the mechanical properties of cementitious composites. Panzera et al.[5] have developed hybrid composites, for restoration works, made of Portland cement, steatite particles and unidirectional carbon fibres, aiming to increase the flexural strength of the material. Cota et al.[6] have studied the mechanical behaviour of composites manufactured with steatite residues and matrices consisting of mixtures of epoxy polymer and Portland cement paste in different proportions. The maximum amount of steatite particles added by these researchers was 60 wt.%. The main difficulties encountered were the low workability and the low capacity of material densification and the loss of mechanical strength, which have been the main limiting factors for an increase of the weight percent of dispersed phase in these composites.

A small number of studies have been carried out with the aim of developing and characterising materials for restoration of historical monuments manufactured with other types of rock. Torney et al.[7] developed a study in order to compare some physical properties of the sandstone with the properties of two types of commercial mortars applied in restoration of rocks. Stefanidou et al.[8] have studied the development of mortar for application in restoration of monuments manufactured with different types of rock. The authors carried out a study to investigate the colour ratio of the mortars according to the aggregate used. It has been found that a correct selection of the type, size and weight fraction of the aggregate can generate artificial materials with colouration and texture similar to natural rock. Particle size and the weight percent of steatite particles incorporated into the composites are the main factors associated with the degree of physical similarity between the artificial material and the rock. Therefore, in order to produce materials with colour and texture similar to soapstone, a high weight percent of dispersed phase in the composite material is desired. This can be achieved by maximising the packing density of steatite particles, a factor that also contributes to increased mechanical strength of the material[9]. Particle packing is the problem of the correct selection of proportions and sizes of the particulate materials. The highest particle packing is obtained when large voids are filled with small particles whose voids are again filled with even smaller particles and so on[10].

Particle packing can be represented quantitatively by the packing density, defined as the mass of solid in a unit of total volume. The materials with high packing density have low volume of voids between particles and, consequently, require a smaller weight percent of binding material as matrix. Various properties of particulate composites are related to the packing of particles representing the dispersed phase. High packing densities are fundamental to obtain particulate composites of low porosity and, in consequence, with enhanced mechanical properties[11].

The particle size distribution of the system or the filling sequence of the voids between the particles determines whether a high packing density is achieved. Therefore, it is important to optimise the relative amounts of different sized particle fractions that constitute the system[12]. This optimisation can be performed by the use of a statistical methodology based on mixture design experiments. Experiments with mixtures are those where two or more ingredients are mixed to form a product. The response of interest to be measured constitutes a property of the mixture, depending only on the proportions of the components present (whether in mass, volume or molar ratio), and not on the total amount of the mixture. A general objective of

DOI: https://doi.org/10.30564/jcr.v1i1.152

this experimental design is the modelling and analysis of the response surface of the mixture. This methodology is based on a limited number of observations considering pre-selected proportions of the components, resulting in mixtures of different compositions[13]. By modelling the surface of response of the mixtures it is possible to obtain a prediction of the response for any composition of the mixture within the region covered by the experiment, and consequently, to estimate the optimum composition of the studied mixture[14].

This work aims to maximise the packing density of steatite particles in order to increase the weight percent of dispersed phase incorporated in composites destined to the restoration of artefacts made of soapstone. This was achieved by optimising the particle size distribution using a statistical methodology based on the planning of three component mixture experiment.

2. Materials and Methods

An epoxy polymer is used as matrix phase, while particles of soapstone are used as dispersed phase.

2.1 Matrix Phase: Epoxy Resin Polymer

A liquid epoxy resin based on bisphenol A, LY-1316 2BR, and Aradur 2963 curing agent, based on cycloaliphatic amines, are used as matrix material for the investigated composites. This set of resin and hardener are produced by the Huntsman Company (Brazil). The recommended ratio between resin and hardener, as well as some other properties, are provided by the manufacturer, as shown in Table 1.

Table 1. Properties of Araldite LY – 1316 2BR and Hardener Aradur 2963 and the Mixture Thereof (Source: Huntsman Ltd.)

Characteristics	Product	Data
Proportion of mixture (in mass)	Resin	100g
	Hardener	48g
Density at 25°C	Resin	1.0 a 1.05 g/cm³
	Hardener	1.0 g/cm³
	Mixture	1.0 to 1.05 g/cm³
Viscosity at 25°C	Resin	1000 - 1200 mPas
	Hardener	30 – 70 mPas
	Mixture	300 – 400 mPas
Time of use at 25°C	Mixture	35 minutes
Time for demoulding at 25°C	Mixture	8 to 10 hours
Curing time at 25°C	Mixture	7 days

This epoxy system (LY – 1316 2BR/2963) was chosen due to its relatively low viscosity and the combination of suitable properties for the composite manufacturing process. This characteristic is fundamental to ensure a high physical adhesion upon porous surfaces and a high percentage of the dispersed phase (steatite particles) without compromising the workability of the material. Moreover, LY - 1316 2BR/2963 epoxy polymer also provides a combination of important and desirable characteristics for restoration applications such as colourlessness. This set also provides a system for easy handling and curing at room temperature. According to the manufacturer, the final product after curing is rigid and presents an excellent balance between its mechanical, thermal and chemical resistance, associated with low contraction, high adhesion, high cohesion and optimum dimensional stability.

2.2 Dispersed Phase: Steatite Particles

The steatite particles were collected from a local manufacturer of pans and artefacts of soapstone, located in the city of Congonhas (Brazil). The steatite residues were dried in an oven at 110°C for 24 hours. The chemical analysis of the soapstone residue was obtained by X-ray fluorescence spectrometry as shown in Table 2. The high content of silicon oxide (44.78 %) and magnesium oxide (29.24 %) found in the soapstone powder is characteristic of the mineral talc, the main constituent of steatite[15].

Table 2. Composition of Steatite residue.

Compound	Quantity (%)
SiO_2	44.78
Al_2O_3	3.65
Fe_2O_3	8.34
TiO_2	< 0.001
CaO	2.99
MgO	29.24
NaO_2	< 0.001
KO_2	< 0.001
MnO	0.15
P_2O_5	0.01
Loss on ignition	**10.36**

Subsequently, the powders were characterised by its grain size distribution, as recommended by the standards ASTM C136M-14[16] and NBR NM 248-03[17]. An electromagnetic stirrer and a set of standard sieves[18] were used to determine the grain size distribution of the steatite residue from the pans manufacturing process (Table 3).

Table 3. Particle size analysis of the steatite residue.

PARTICLE SIZE DISTRIBUTION - STEATITE (300g)								
SIEVE		SAMPLE 1		SAMPLE 2		% Mean		
ASTM E11-17 [18]	Opening (mm)	Retained mass (g)	% Retained	Retained mass (g)	% Retained	Retained	Cumulative percent retained	Cumulative percent passing
3”	75.00	0.00	0.00	0.00	0.00	0.00	0.00	100.00
1.1/2”	37.50	0.00	0.00	0.00	0.00	0.00	0.00	100.00
3/4”	19.00	0.00	0.00	0.00	0.00	0.00	0.00	100.00
3/8”	9.50	0.00	0.00	0.00	0.00	0.00	0.00	100.00
4	4.75	5.17	1.73	5.37	1.79	1.76	1.76	98.24
8	2.36	22.10	7.38	22.80	7.61	7.50	9.26	90.74
16	1.18	47.20	15.77	47.07	15.72	15.74	25.00	75.00
30	0.60	72.59	24.25	71.79	23.97	24.11	49.10	50.90

 DOI: https://doi.org/10.30564/jcr.v1i1.152

50	0.30	48.74	16.28	48.08	16.05	16.17	65.27	34.73
100	0.15	32.82	10.96	31.53	10.53	10.74	76.01	23.99
PAN		**70.77**	**23.64**	**72.88**	**24.33**	**23.99**	**100.00**	**0.00**
TOTAL		**299.39**	**100.00**	**299.52**	**100.00**	**100.00**	**226.40**	
Fineness modulus: 2.26								

The grain size distribution of the steatite residue is close to the limits established by standards NBR 7211-09[19] and ASTM C144-11[20] but does not fully comply with them. Therefore, to be used as fine aggregate in concretes and mortars, the soapstone residue must have its grain size distribution adjusted to suit the requirements provided by these standards. The grain size distribution curves are shown in Figure 1.

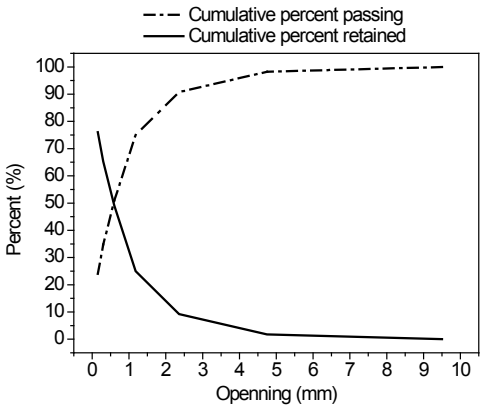


Figure 1. Particle Size Distribution of Steatite Residue

The particle size distribution curve presents a continuous behaviour in the size range between 4.75 mm and 0.15 mm. This behaviour indicates that the investigated steatite residue is well-graded and does not present discontinuities in the analysed range. Aggregates with a continuous particle size distribution favour higher particle packing densities[21].

The particle size distribution of steatite used in this work is obtained following the recommendations of ASTM C136M-14[16] and NBR NM 248-03[17]. However, the grain size range for composite manufacturing is selected prioritising the best material suitability for restorations. Dense particle packing is fundamental to obtain particulate composites with low porosity and, consequently, materials with enhanced mechanical properties. Therefore, an effort is made to use an aggregate with a large and continuous particle size distribution, as these factors contribute positively to the increase of the particle packing density[21]. However, an upper limit and lower limit of particle size are established aiming at a better suitability of the material for its application in restoration works.

Composites produced with particles larger than 1.18 mm showed little penetration capacity into cracks, turning the material unsuitable for application in this type of defect[9]. Composites made with particles smaller than 0.15 mm were also studied. These fine particles were lighter in colour than the natural stone. Consequently, the preliminary composites made of these particles exhibited a very different physical appearance as the rock[9]. Therefore, the lower (0.15 mm) and upper (1.18 mm) limits of the steatite particle size was established. Within the established size range, the steatite is classified in monodisperse particles according to the standards NBR 7211-09[19] and ASTM C144-11[20]. These monodisperse particles are denominated coarse, medium and fine, as presented in Table 4 and in Figure 2.

Table 4. Monodisperse Particle Levels of Steatite (ASTM E11-17[18])

Particle Size	Sieve passing	Sieve retained
Coarse	16	30
Medium	30	50
Fine	50	100

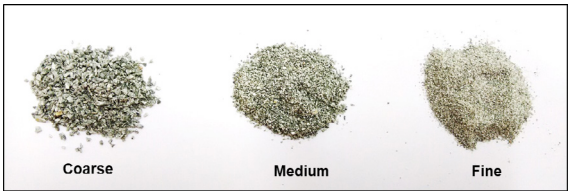
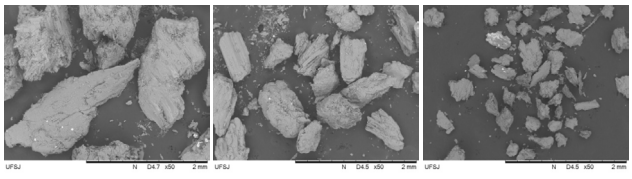


Figure 2. Steatite Classified in Coarse, Medium and Fine Monodispersed Particles.

The characterisation of the steatite particle morphology was performed by scanning electron microscopy (SEM), Hitachi MEV-TM 3000, using backscattered electron mode (BSE) at 15 kV and carbon tape. Figure 3(a-c) shows the images of steatite particles at 50× of magnification.



(From Left) Figure 3. (a) Coarse (1.18 – 0.60 mm), (b) medium (0.60-0.30 mm) and (c) fine sized (0.30 – 0.15 mm) steatite particles.

The SEM images were compared with the standard NBR 7389-09[22] to define the levels of sphericity and roundness of the particles. Through visual evaluation a great heterogeneity in the particle shape is identified, with

DOI: https://doi.org/10.30564/jcr.v1i1.152

particles of low sphericity and angular and subangular surfaces showing little evidence of wear. This morphology is characteristic of aggregates that undergo artificial comminution processes[23], as is the case of the soapstone particles used in this work. This type of morphology presented by the soapstone particles contributes negatively to the packaging. The further away from the spherical and angular shape are the particles, the lower the packing density of a distribution containing them. This is due to friction effects between particles with irregular surfaces. The smaller the size of the irregular particles, the greater this effect will be, due to the greater specific surface area[21].

2.3 Experimental Planning with Mixtures

In order to maximise the packing density of the steatite particles, the mixture design of experiment was used in this project. The simplex-centroid design for mixtures with three components was applied in the system consisting of coarse, medium and fine monodispersed particles. This model is represented by an equilateral triangle with seven points; the dots represent the different pre-selected compositions that are tested. In a three-component mixture, each experiment at the vertices represents the formulations of a pure component, while the centre of the edges of the equilateral triangle represents the binary mixtures and compositions within the triangle the ternary mixtures, as shown in Figure 4.

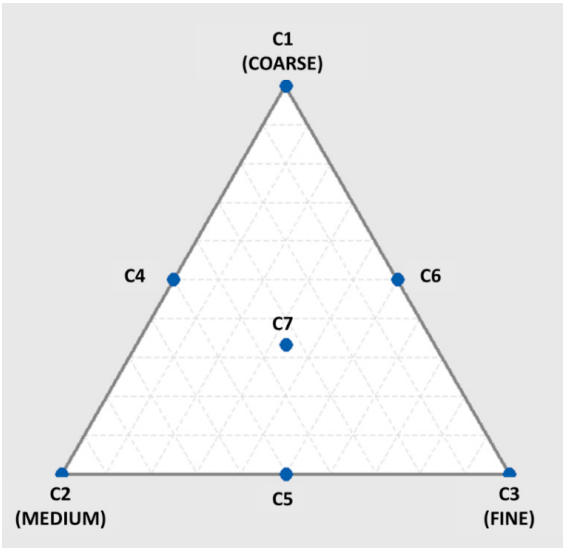


Figure 4. Simplex-centroid Surface for Three Distinct Particle Size Classes

The components of each composition were weighed and mixed to homogenise the material. Subsequently, one at a time, the mixtures were placed in a graduated cylinder and subjected to vibration on an electromagnetic stirrer for three minutes. The packing density of each composition is calculated by dividing the mass of each mixture by the total volume occupied by the particles in the beaker after vibration. This procedure was performed for the seven different compositions and repeated twice, in order to minimise the experimental error of the results[24]. The randomisation method was adopted during conduction of the tests, allowing an arbitrary ordering of the different compositions and avoiding that effects of uncontrolled factors could affect the response-variable[25][26].

The values obtained in the tests with pre-selected compositions were used to determine the coefficients (β) of the canonical polynomials of Scheffé and to generate the response surface (using Minitab software version 17, module "Mixture")[27].

Two polynomial models applicable to the simplex-centroid design of three components (q = 3) were tested: quadratic model (Equation 1) and special cubic model (Equation 2).

$$\hat{y} = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \quad (1)$$

$$\hat{y} = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 + \beta_{123} x_1 x_2 x_3 \quad (2)$$

On the equations 1 and 2, $\hat{y}$ is the expected response for the proposed mixture, and x_1, x_2 and x_3 represent the proportion of each of the three components of the mixture. The parameters $\beta_i x_i$ represent the expected response for the pure compositions. The terms $\beta_{ij}\, x_i\, x_j$ represent an alteration of the quadratic model response (degree two) over the linear model (degree one). The third-degree term $\beta_{ijk} x_i\, x_j\, x_k$ describes the response surface behaviour in the interior of the surface simplex[28]. After the experimental packing tests, the polynomial model (R^2 and R^2 adjusted) that presented the best fit is chosen, and consequently, the best predictive capability for the response variable investigated.

The adjusted equation relates the packing density of the particles to the proportions of the three monodisperse particles used. By processing of the experimental data, a response surface was generated for the variable packing density. By means of the adjusted equation and the generated response surface it was possible to predict the behaviour of the response variable for different combinations of the monodisperse particles used. Thus, the packing density could be maximised by optimising the amounts of the three distinct monodisperse particles of different size ranges through non-linear programming.

2.4 Sample Preparation

Composite samples with the highest possible weight percent of dispersed phase were manufactured after the optimisation of the amounts of the three monodisperse steatite particles, aiming at the development of a material with similar colour and texture as steatite rock. Based on

 DOI: https://doi.org/10.30564/jcr.v1i1.152

previous studies[4][5][6] and preliminary tests, composites were produced using three different weight percent of matrix phase (35 wt.%, 30 wt.% and 25 wt.%) and dispersed phase (65 wt.%, 70 wt.% and 75 wt.%).

The composites were manufactured by manually mixing 100 parts by mass of resin and 48 parts by weight of hardener for a period of 3 minutes, following the manufacturer's recommendations. After homogenisation of the resin with the hardener, the polymer phase was manually mixed with the steatite particles in the different proportions over a period of 3 minutes. The materials were cast in silicone moulds and subjected to vibration in an electromagnetic stirrer for material accommodation and elimination of air bubbles entrapped. A mould with cylindrical shapes with dimensions recommended by ASTM D695-10[29] is shown in Figure 5.

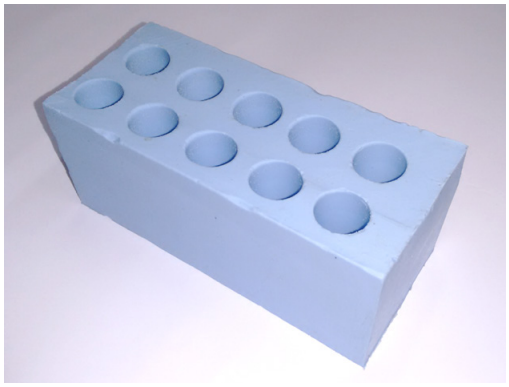


Figure 5. Silicon Mould Used to Manufacture Sample

After 48 hours the specimens of 40 mm height and 20 mm diameter were removed from the mould, identified and packed in a sealed plastic container. Curing of the specimens was performed at room temperature for a period of 7 days. Ten specimens from each of the three compositions were fabricated in a randomised order. In total, 30 specimens were produced, as recommended by ASTM D695-10[29].

3. Results

The simplex-centroid design for mixtures with three components was applied to the system consisting of coarse, medium and fine monodisperse particles. The packing densities of the seven pre-selected mixtures by the simplex-centroid design are shown in Table 5.

Table 5. Packing Density of the Pre-selected Mixtures of Steatite Particles

Compositions	Packing density (g/cm³)			
	R1	R2	Mean	Standard deviation
C1	1.420	1.429	1.425	0.006
C2	1.337	1.351	1.344	0.010
C3	1.238	1.225	1.232	0.009
C4	1.453	1.471	1.462	0.012
C5	1.488	1.479	1.484	0.006
C6	1.374	1.359	1.366	0.011
C7	1.471	1.479	1.475	0.006

The values obtained in the tests with pre-selected mixtures were used to determine the coefficients of the canonical polynomials of Scheffé and to generate the response surface for the variable packing density (using Minitab software version 17, module "Mixture"). Two polynomial models applicable to the three-component simplex-centroid delineation (q = 3) were tested, the quadratic model (Equation 1) and the special cubic model (Equation 2).

The quadratic model was chosen because it presented a better adjustment capacity to the experimental data (R^2 = 99.44 % and R^2 adjusted = 99.10 %). The adjusted R^2 and R^2 values close to 100 % indicate that the quality of the fit of the model is satisfactory. The adequacy of the statistical model used is verified through the normal probability plot for the residues presented in Figure 6. The behaviour of the residuals for the response variable packaging density of steatite particles was adequate for normal distribution conditions. The points are located approximately along a straight line and there are no outliers, which are points distant from the line and may represent a source of error in the data collection.

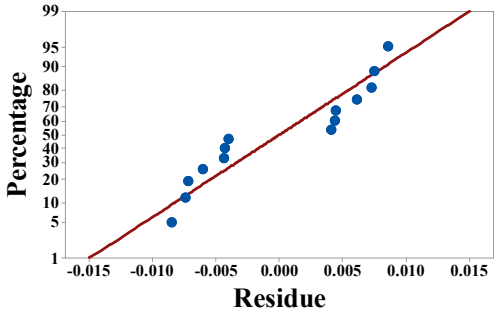


Figure 6. Normal Residual Probability Plot for the Packing Density of Steatite Particles

After the definition of the polynomial model and the verification of the adequacy of the model, the coefficients obtained through the statistical planning and the experimental data were inserted in Equation 1. The quadratic polynomial adjusted to the experimental data of packaging density of the steatite particles is presented in Equation 3.

$$\hat{y} = 1.42x_1 + 1.34x_2 + 1.23x_3 + 0.31x_1x_2 + 0.63x_1x_3 + 0.32x_2x_3 \quad (3)$$

Equation 3 relates the variable response packing density of the particles ($\hat{y}$) with the proportions of the monodisperse coarse (x_1), medium (x_2) and fine (x_3) particles used. By adjustment of the equation to the experimental data it is possible to predict the behaviour of the variable response for all different combinations of the monodisperse mixtures. Consequently, it was possible to maximise the packing density of the particles by optimising the particle size distribution in the range investigated.

DOI: https://doi.org/10.30564/jcr.v1i1.152

The theoretical proportions of monodisperse particles for maximum packing density were obtained by maximising Equation 3.

The maximum particle packing density was obtained for a ternary mixture, composed of 62% of coarse particles (1.18 mm - 0.60 mm), 6% of medium sized particles (0.60 mm - 0.30 mm) and 32% of fine particles (0.30 mm - 0.15 mm). Inserting the values of the proportions in Equation 3, a theoretical packing density of 1.50 g/cm³ was obtained. This value was verified and confirmed by performing new experimental tests using the statistical modelling data.

Through experimental design with mixtures it was also possible to generate a contour plot for the behaviour of the variable packing density for all different combinations of proportions of the coarse, medium and fine monodisperse particles, see Figure 7.

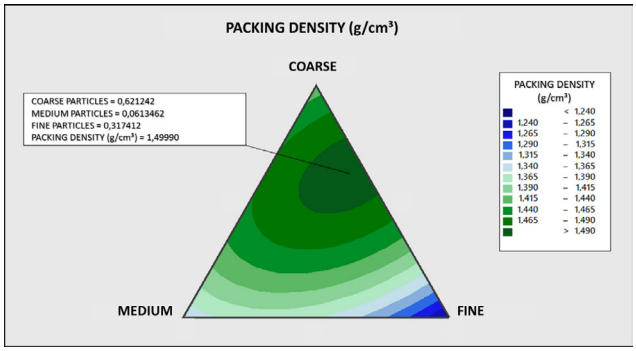


Figure 7. Contour Plot for the Variable Packing Density

Based on the contour plot it is possible to estimate the region of the experimental space which maximises packing density response of the steatite particles. As shown in Figure 7, the coarse monodispersed particles presented a higher packing density compared to medium and fine monodisperse particles, however, the region of the experimental space that presents the highest packing density is a ternary mixture composed of coarse, medium and fine particles. These findings confirm the theories reported in previous work[21], assuming that larger irregular particles with a continuous particle size distribution produces denser packages.

After the optimisation of the size distribution of the steatite particles, composites were fabricated using an epoxy polymer (Table 1) as matrix and different weight percent of dispersed phase (65 w.t%, 70 w.t% and 75 w.t%) with the optimised grain size distribution.

In the attempt to develop a material with aesthetic characteristics similar to those presented by soapstone, the largest possible weight percent of dispersed phase was incorporated to the composites. Therefore, based on previous studies[4][5][6] and preliminary tests, composites with 25 wt.% of epoxy polymer and 75 wt.% of steatite particles were manufactured. However, this material presented very low fluidity, compromising the workability and the capacity of densification, leading to a large amount of external macro pores (Figure 8), especially for a cylindrical sample that makes it difficult to remove trapped air through its longitudinal length.

Due to the low fluidity of these composites, which resulted in high porosity, the weight percent of polymer matrix was increased, and the weight percent of steatite particles decreased. Thus, composites were fabricated with 70 wt.% and 65 wt.% of steatite particles. Contrary to the first percentage used, the mixture composed of 30 wt.% epoxy polymer and 70 wt.% of steatite particles revealed better workability and higher density with reduced amount and size of external macro pores. The blend consisting of 35 wt.% epoxy polymer and 65 wt.% steatite particles also showed adequate workability and density. However, this higher percentage of epoxy polymer (35 wt.%) was excessive and caused segregation of the steatite particles. The dispersed phase accumulated predominantly in the lower part of the specimens, generating a heterogeneous material along the longitudinal section (Figure 8).

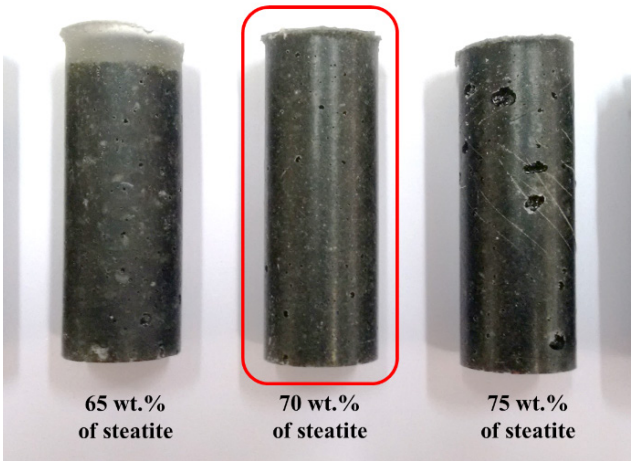
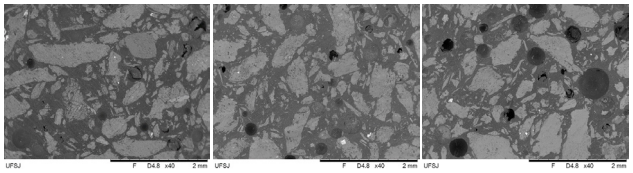


Figure 8. Polymer-based Composites with 65 wt.%, 70 wt.% and 75 wt.% of Steatite Inclusions

The Hitachi MEV-TM 3000 Scanning Electron Microscope operating with backscattered electron mode at 15kV was used for the microstructural analysis of the composites. Figure 9 shows the composite images containing 65 wt.% (a), 70 wt.% (b) and 75 wt.% (c) of steatite particles with a 40 × magnification. Composites made with a large amount of steatite particles reveal the presence of a larger amount of macro pores (Figure 9c), which can be attributed to the different rheology of the system that can affect the removal of internal bubbles. A good interface condition between the steatite particles and the matrix phase is evidenced even for the inclusion of large amounts of par-

 DOI: https://doi.org/10.30564/jcr.v1i1.152

ticles (75 wt.%), without pore formation or discontinuity.



(From Left) Figure 9. Polymer-based Composites with 65 wt.% (a), 70 wt.% (b) and 75 wt.% (c) of Steatite Particles

Table 6 shows bulk density values for the soapstone (natural stone) and composite materials. The bulk density of the composites ranged from 1.753 g/cm³ to 1.828 g/cm³. The composites obtained lower bulk density in relation to the natural soap stone due to the low-density polymer matrix (1.05 g/cm^3, Table 1). The composites made with 30 wt.% of polymer and 70 wt.% of dispersed phase reached the highest mean bulk density due to their lower amount of matrix phase and internal pores. In addition, composites made with 30 wt.% of polymer matrix and 70 wt.% of steatite particles also exhibited colouration and texture very similar to natural rock as shown in Figure 10.

Table 6. Bulk Density of Natural Steatite Rock and Composite Materials

Compositions	Bulk density (g/cm³)	
	Mean	Standard deviation
Steatite (soapstone)	2.884	0.005
C1 (35 wt.% of polymer and 65 wt.% of steatite)	1.753	0.009
C2 (30 wt.% of polymer and 70 wt.% of steatite)	1.828	0.007
C3 (25 wt.% of polymer and 75 wt.% of steatite)	1.802	0.011

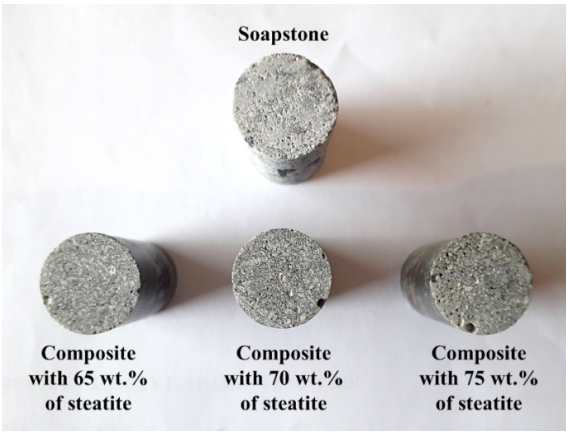


Figure 10. Aesthetic Characteristics of Soapstone and Composite Materials

Finally, composites made with 30 wt.% of polymer matrix and 70 wt.% of dispersed phase reached the most suitable properties and aesthetic characteristics for the restoration of soapstone structures. In previous works[4][5][6], the maximum weight percentage of steatite particles in composites for restoration proposal was 60 wt.%.

4. Conclusion

The statistical methodology based on the three-component mixture experiment was used to optimise the particle size distribution of steatite particles. By means of this technique, the packing density of the steatite particles was maximised (1.50 g/cm³) by the optimisation of the size distribution within the investigated range. The optimised distribution allowed the production of composites with 70 wt.% of dispersed phase, 10% higher than the highest percentage used in previous works. This high percentage of weight contributes positively to the development of materials destined to the restoration of soapstone structures, with aesthetic characteristics similar to those of rock.

Acknowledgements

The authors would like to thank the Brazilian Research Agencies, CNPq (PP & PDE-205255/2017-5) and FAPEMIG (PPM-00075-17) and CAPES (MSc scholarship), for the financial support provided.

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