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An elastomeric material for facial prostheses: synthesis, experimental and numerical testing aspects

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Abstract

Current materials used for maxillofacial prostheses are far from ideal and there is a need for new improved materials which better simulate the tissues they are replacing. This study was based on a mixed experimental/analytical/numerical approach. A new polymeric material was developed to provide a better alternative to the materials currently used in maxillofacial prosthetics. A series of experimental tensile tests were performed in order to characterise the tensile properties of the material. A Mooney–Rivlin type hyperelastic formulation was chosen to describe the constitutive behaviour of the polymer which operates at the finite strain regime. The material parameters (two) of the constitutive law were identified with the experimental data. The Mooney–Rivlin material was found to be suitable to represent accurately the mechanical behaviour of the polymer up to 50% strain as shown by the excellent agreement between analytical and experimental results. An FE model reproducing all the characteristics of the experimental tensile tests was built and a series of three FE analyses were conducted and has proven the proper finite element implementation of the material model. This preliminary study will serve as a basis to introduce more complex features such as viscoelasticity and wrinkling of the soft polymeric structure in order to optimise the performances of the final prosthetic material.

Keywords: Maxillofacial prosthetics; Silicone rubber; Mooney–Rivlin; Finite element

1. Introduction

Maxillofacial materials are used to replace missing facial parts which have been lost through disease or trauma. They are usually comprised of poly(dimethylsiloxane) (PDMS) elastomers. Although widely used, these materials are far from ideal. The quality of these materials depends greatly on their two basic components, the PDMS chains and the silica fillers, and the interactions between these affects the overall strength and service life of the material. Important properties essential in a material used for the construction of maxillofacial prostheses are high tear strength, low hardness and a low enough viscosity to make manipulation of the uncured material manageable [1–4].

It is also imperative that facial tissue substitutes mimic as closely as possible the natural behaviour of facial soft tissues. In order to bring a substantial improvement in this matter, a very soft new three-layered polymeric system is currently under development at the Matrix Biology and Tissue repair Research Unit in Cardiff. The silicone rubber material used in this study was designed to be the base material of the novel three-layered system which will also include an inner silicone gel and a thin outer polymeric coating.

The mechanical properties of a silicone elastomer are dependent on many factors. An important one of these is the molecular weight distribution which affects the mechanical properties of the elastomer. The blending of long and short chains of the same polymer gives a broader, bimodal molecular weight distribution, and a network prepared from such a blend is known as a bimodal network [5]. The practical significance of such networks is to achieve elastomers possessing a combination of good mechanical properties such as tear strength, tensile behaviour and resilience [6–8].

A second factor affecting mechanical properties is the incorporation of a hydrophobic surface treated silica

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filler (with dimethyl silyl or trimethyl silyl groups) with a low particle size and therefore a high surface area. This prevents incorporation of water and increases the strength of the elastomer. Clinically acceptable mechanical properties are achieved only at the correct filler concentration [9].

The third main factor is the degree of cross-linking between polymer chains. A high cross-link density produces an inelastic brittle material whilst a very low cross-link density would produce a very weak material with low tear and tensile strength [9].

In the present study the percentage ratio of low molecular weight polymer to high molecular weight polymer was varied to give a range of biomodal formulations. In addition, the amount of surface treated filler was varied whilst keeping the cross-linker level constant.

Finite element analysis (FEA) is a computer modelling method which enables the accurate simulation and prediction of material responses. FEA modelling techniques will be used to facilitate in the engineering of the new silicone materials.

The initial objective of this study was to develop a new maxillofacial silicone rubber material with superior properties in comparison to commonly used commercially available materials. A further objective was to perform simple tensile tests at different loading rates on the new silicone rubber base material, to propose a suitable constitutive formulation and validate its proper implementation into a commercial finite element code.

2. Materials and methods

2.1. Silicone rubber formulation

The constituents of the silicone rubber formulation used in this study are shown in Table 1. The four formulations developed are shown in Table 2. The silicone rubber material was prepared by first blending the high and low molecular weight vinyl end blocked polymers. The silica filler and hydride functional filler were then added and the polymeric base was mixed for 2 h. Platinum catalyst was added at 0.15% w/w and the material was cured at 100°C for 1 h. The four formulations tested were compared to two widely used commercially available materials, Factor II A-2186 (Factor II Inc., Lakeside, Ca, USA) and Technovent Silicone Elastomer (Technovent Ltd., UK).

2.2. Hardness test

Five hardness specimens (45 mm × 45 mm × 4 mm) were made from each material using a conventional dental flasking technique. The hardness test was conducted according to A.S.T.M. D-1415 (1983) [10] using a Wallace Dead Load Hardness Tester (H.W. Wallace and Co. Ltd., Croydon, UK) with results being read directly in International Rubber Hardness Degrees (IRHD). For each specimen, 10 hardness readings were measured at 10 different positions on the surface of the specimen.

2.3. Tear test

Test specimens (100 mm × 80 mm and 2 mm) were made using the conventional dental flasking technique. Ten tear specimens were cut from the processed material with final dimensions of 50 mm × 10 mm × 2 mm, with a 4 mm cut placed from one edge. This test specimen is a modification of the test specimen described by ASTM D624: Standard Test Method for Rubber Property—Tear resistance [11].

Testing was carried out using the Lloyd Instruments LR 10K testing machine at a constant crosshead speed of 20 mm/min. Tear strength was calculated using the following equation:

$$T_s = F/t,$$

where T_s is the tear resistance (N/mm), F the load at failure (N), t the thickness of specimen (mm).

2.4. Viscosity test

Degassed silicone (25 g) was placed in a universal tube and clamped under the viscometer with the spindle immersed within the sample.

The test was carried out using a Brookfield DV-I+ Viscometer (CP Instrument Company Ltd., Herts, England) fitted with the RV7 spindle. The spindle was driven at three chosen speeds (0.5, 1.0 and 2.5 rpm)

Table 1
Constituents of silicone rubber formulation

Materials	Description	Manufacturer
PDMS base polymer (V46)	High molecular weight vinyl end blocked poly(dimethylsiloxane)	ABCR, Manchester, UK
PDMS base polymer (V21)	Low molecular weight vinyl end blocked poly(dimethylsiloxane)	ABCR, Manchester, UK
Filler	Surface treated hydrophobic silica	DegussA Ltd., Cheshire, UK
Cross- linker	Hydride functional silicone polymer	ABCR, Manchester, UK
Catalyst	Platinum complex	ABCR, Manchester, UK

Table 2
Bimodal formulations developed

Formulation	Polymer ratios	Polymer content	Filler content
1	V46 80%, V21 20%	70	30 (ABCR)
2	V46 85%, V21 15%	75	25 (ABCR)
3	V46 95%, V21 5%	75	25 (ABCR)
4	V46 80%, V21 20%	60	40 (ABCR)

within the test fluid. Results were read off the viscometer face and measured in units of centipoise (cP) or millipascal seconds (mPa s).

2.5. Tensile testing of optimum formulation

Formulation 2 was deemed to have the best compromise of tear hardness and viscosity properties and therefore further tensile testing was performed on this formulation. Three identical dumb-bell shaped specimens in accordance with BS 903 (1979) type 2 test pieces [12] were cut from strips of processed polymeric material. The specimens were tested according to BS 903 Part A2 (1979)—Tensile Stress Strain Properties [12].

Elongation tests were carried out on a Lloyd Instruments LR10K tensile machine. Stress–strain curves were obtained by at constant cross head speeds of 0.2, 2 and 6 mm/min (Table 3).

2.6. Processing of raw experimental data

The displacement–force elongation curves obtained experimentally for each loading rate were converted into stretch–nominal stress curves for further identification. A Mooney–Rivlin type formulation was chosen for the constitutive modelling of the new polymeric material [13]. This formulation has been widely used in rubber and polymer elasticity because it accommodates well the non-linear characteristics of these materials. As the polymer tested undergoes negligible volume changes at low and finite strain regime the common hypothesis of incompressibility was made. This also facilitates the identification process when out-of-plane deformations need to be known.

2.7. Constitutive law

An incompressible Mooney–Rivlin material is an isotropic hyperelastic material whose mechanical behaviour is characterised by a strain energy function ψ of the following form:

$$\psi = C_1(I_1 - 3) + C_2(I_2 - 3) + \frac{\kappa}{2}(I_3 - 1)^2, \tag{1}$$

Table 3
Mooney–Rivlin material coefficients identified for each elongation rate

Elongation rate (mm/min)	0.2	2	6
C_1 (kPa)	90.35	57.65	48.37
C_2 (kPa)	12.82	50.15	80.17

where I_1, I_2 and I_3 are the first principal invariants of the right and left Cauchy–Green deformation tensors $\mathbf{C}$ and $\mathbf{b}$. C_1, C_2 are material parameters while ψ is a Lagrangean multiplier enforcing the incompressibility constraint. The Lagrangean stress tensor, $\mathbf{S}$, known as the second Piola–Kirchhoff stress tensor is obtained by first differentiation of ψ with respect to $\mathbf{C}$:

$$\begin{aligned}\mathbf{S} &= 2\frac{\partial\psi}{\partial\mathbf{C}} \\ &= 2\left[\left(\frac{\partial\psi}{\partial I_1} + I_1\frac{\partial\psi}{\partial I_2}\right)\mathbf{1} - \frac{\partial\psi}{\partial I_2}\mathbf{C}\right] + p\mathbf{C}^{-1}.\end{aligned} \tag{2}$$

The nominal stress tensor or first Piola–Kirchhoff stress tensor $\mathbf{P}$ is easily obtained from $\mathbf{S}$ by means of the simple tensorial transformation: $\mathbf{P} = \mathbf{S}\mathbf{F}^{\mathrm{T}}$, where $\mathbf{F}$ is the deformation gradient and the superscript T means transpose.

$$\mathbf{P} = 2\left[\left(\frac{\partial\psi}{\partial I_1} + I_1\frac{\partial\psi}{\partial I_2}\right)\mathbf{F}^{\mathrm{T}} - \frac{\partial\psi}{\partial I_2}\mathbf{C}\mathbf{F}^{\mathrm{T}}\right] + p\mathbf{F}^{-1}. \tag{3}$$

The expression of the Cauchy stress tensor is obtained by simple push-forward operation on $\mathbf{S}$ (in the particular case of an incompressible material, J, the Jacobian of the deformation is equal to unity):

$$\begin{aligned}\sigma &= \frac{1}{J}\mathbf{F}\mathbf{S}\mathbf{F}^{\mathrm{T}} \\ &= 2\left[\left(\frac{\partial\psi}{\partial I_1} + I_1\frac{\partial\psi}{\partial I_2}\right)\mathbf{b} - \frac{\partial\psi}{\partial I_2}\mathbf{b}^2\right] + p\mathbf{1}.\end{aligned} \tag{4}$$

where $\mathbf{1}$ is the second-order unit tensor.

In a uniaxial deformation mode the stretch in the tensile direction λ_u is related to the two other principal stretches, λ_2 and λ_3 by the incompressibility constraint:

$$\lambda_u\lambda_2\lambda_3 = 1 \Rightarrow \lambda_2 = \lambda_3 = 1/\sqrt{\lambda_u}. \tag{5}$$

The nominal tensile stress T is expressed as follows:

$$T = \left(1 - \frac{1}{\sqrt[3]{\lambda_u}}\right)(C_1\lambda_u + C_2). \tag{6}$$

2.8. Identification

Using a simple least-squares-fit procedure minimising the relative difference ε between the n nominal stress–nominal strain data pairs of the experimental (e) and analytical (a) tensile tests, the material coefficients C_1

and C_2 were determined:

$$\varepsilon = \sum_{i=1}^{n} \left(1 - \frac{T_i^{a}}{T_i^{e}}\right)^2. \quad (7)$$

The identification process was conducted for data pairs corresponding to strains of up to 100%, but given that the prosthetic material is unlikely to undergo such deformations in vivo, the final identified coefficients were established for a maximum strain of 50% (Table 1). Excellent agreement was obtained between the theoretical and experimental results for the three material coefficients sets (Figs. 1–3).

2.9. Finite element analyses

For each sample tested a corresponding solid model was built and meshed with hexahedral 8-noded elements (C3D8) within the pre-processing software ABAQUS/CAE (HKS Inc., Pawtucket, RI, USA). Each solid model represented the geometry of the dumb-bell shaped specimen where the deformations and the stresses are assumed to be uniform. This therefore excluded the gripped part of the specimen. The same fine mesh density was used for the three models (3294 nodes and 2400 elements). Three non-linear FE analyses reproducing the exact characteristics of each tensile test (state of homogeneous deformations) were carried out with the implicit ABAQUS solver. Material coefficients obtained by previous identification were used as input for the incompressible Mooney–Rivlin material implemented as standard within the FE code. As no significant kinematic constraints other than incompressibility were present within the material (because of the state of homogeneous deformations), no need was found to use hybrid element formulation that prevents the well-known "locking" phenomenon appearing in FE analyses of isochoric media [14].

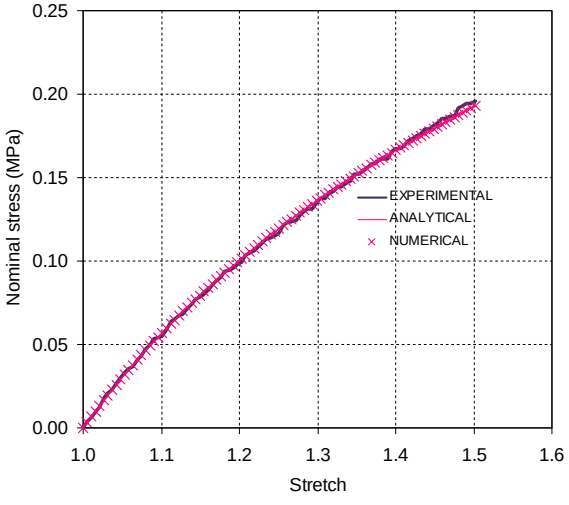

Fig. 1. Tensile curves for the 0.2 mm/min elongation rate.

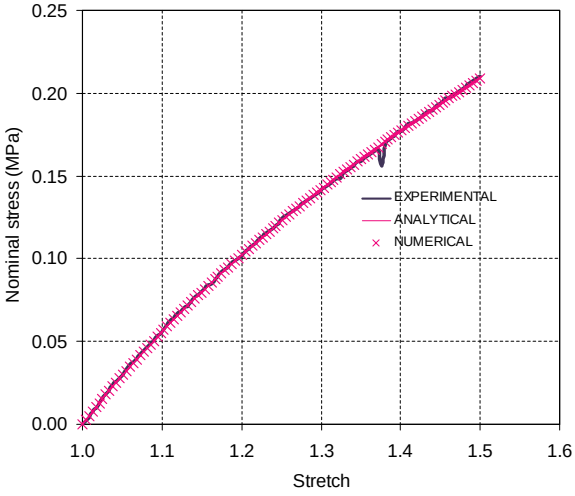

Fig. 2. Tensile curves for the 2 mm/min elongation rate.

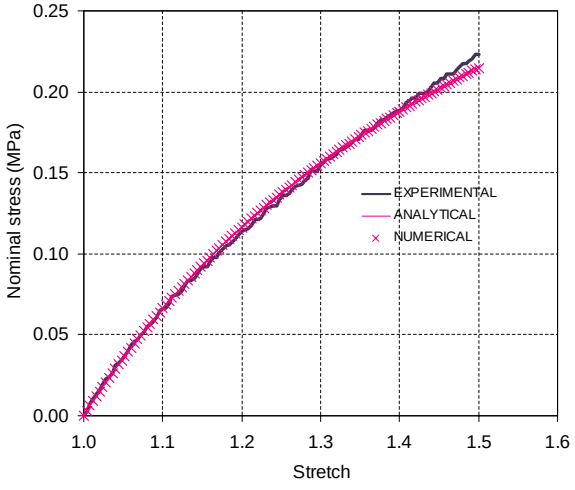

Fig. 3. Tensile curves for the 6 mm/min elongation rate.

2.10. Statistics

One-way analysis of variance (ANOVA) was used to test for any significant difference between the mean values of the materials tested. Post-tests (Bonferroni method) were used to determine whether the mean value of any particular material differed significantly from another specified material, while considering all the data.

3. Results

The hardness results for the four experimental formulations in comparison to the commercial materials are shown in Table 4. Formulations 3 and 4 had significantly higher hardnesses in comparison to formulations 1 and 2 and the two commercially available

Table 4
Tear strength, hardness and visciosity (uncured) of the experimental formulations in comparison to the commercial materials

Material	Tear (N/mm) (mean ± SD)	Hardness (IRHD) (mean ± SD)	Viscosity (C_p)
Formulation 1	6.89 ± 0.75	13.81 ± 0.35	424×10^3
Formulation 2	12.76 ± 1.24	19.35 ± 0.34	200×10^3
Formulation 3	24.28 ± 2.89	33.25 ± 0.26	468×10^3
Formulation 4	16.59 ± 2.24	36.45 ± 0.36	528×10^3
Technovent	1.41 ± 0.26	15.31 ± 0.42	256×10^3
Factor II	5.58 ± 0.35	18.25 ± 1.38	116×10^3

materials ($p < 0.001$). There was no significant differences in the hardness values for formulations 1 and 2 and the two commercially available materials.

The tear strength results for the four experimental formulations in comparison to the commercial materials are shown in Table 4. Formulation 3 had a significantly higher tear strength in comparison to all other materials ($p < 0.001$) and formulations 2 and 4 also had tear strengths significantly higher than the two commercially available materials ($p < 0.001$).

The viscosity results for the four experimental formulations in comparison to the commercial materials are shown in Table 4. Formulation 4 and the Factor II material had significantly higher viscosities in comparison to the other materials ($p < 0.001$). There was no significant difference in the viscosity of all the other materials.

Based on the above results, formulation 2 was deemed to have the best compromise of properties and therefore tensile testing was carried out which was the basis for the numerical analysis.

Experimental, predicted analytical and numerical (FE) nominal stress–stretch curves for formulation 2 are represented in Figs. 1–3 and correspond to the tests performed, respectively, at the following elongation rates: 0.2, 2 and 6 mm/min. Excellent match between the numerical and theoretical results demonstrates the validity of the FE methodology adopted (in addition to the proper implementation of the Mooney–Rivlin material model). The stiffening characteristics of the material as the loading rate increases are clearly exhibited on the stress–stretch curves.

4. Discussion

Maxillofacial prostheses play a crucial role in the rehabilitation of patients who have suffered severe facial disfigurement. Relatively little work has been done on the development of new improved materials and currently used materials have remained virtually the same since the introduction of silicone rubber in the 1950s.

In this work a new silicone rubber maxillofacial material has been developed, mechanically tested and results are analysed via FEA. The material is the base material of a unique new three-layered maxillofacial prosthetic system. The study was based on a mixed experimental/analytical/numerical approach and will be the foundation for more complex studies on the developing three-layered system.

In developing the new base material tear strength, hardness and viscosity were used to assess which formulation went onto further numerical evaluation. It is important that facial prostheses have adequate thin edged tear resistance and are soft enough to go some way toward mimicking facial skin. In addition, the viscosity of the uncured material must be low enough for easy manipulation and colouring by the maxillofacial technologist.

The results show that by altering the ratios of low molecular weight to high molecular weight polymer, it is possible to optimise the tear strength. This is due to the addition of the low molecular weight polymer producing a broader molecular weight distribution and resulting in a local high cross-link density (due to the short chain DMS V21) between relatively long chain DMS V46 polymer. The extra cross-links from the low molecular weight polymer tighten the cross-link network whilst at the same time the flexibility of the longer chains is retained. The results show that at low bimodal concentrations the tear strength is optimised and at higher concentrations the tear strength is reduced. This is because at higher concentrations of low molecular weight polymer, the cross-link network is tightened to such an extent that the flexibility is reduced and more brittle specimens with reduced tear strength are produced.

The highest tear strength formulations however also had high hardness in comparison to the commercial materials. Formualtion 2 offered a good compromise of properties with adequate tear strength, a similar hardness to the commercial materials and a relatively low viscosity. This formulation was then the basis of the further tensile tests and the numerical analysis.

The Mooney–Rivlin constitutive material model has been shown to be suitable to represent accurately the tensile behaviour of the new polymeric materials at constant loading rates. The numerical model was validated by providing excellent agreement between

experimental and analytical results. Viscoelastic effects, highlighted in this study by the difference in the mechanical response according to the loading rate, that may be relevant in maxillofacial prosthetics, were not taken into account but will be in the next phase of this preliminary study. A discrete relaxation function will be introduced by means of a Prony series expansion and appropriate material tests will be designed to allow for the identification of a hyperviscoelastic constitutive law. Wrinkling behaviour of the soft tissue substitute is sought and it is believed that it is the key for offering a human-like realism.

FEA of wrinkling structures are still challenging and particular computational techniques need to be adapted or developed for these very specialised purposes. Each of the three layers composing the new artificial tissue will be analysed separately and after this stage, a global FE model will be built in order to carry out structural optimisation. The optimisation criteria will be related to the mechanical ability to mimic the natural behaviour of facial skin, i.e., the capacity to wrinkle under very small in-plane compressive stresses. To achieve these optimisation objectives alteration of the structural arrangement of the multi-layered material will be considered.

5. Conclusion

A silicone rubber formulation has been developed which will be the base of a novel new three-layered maxillofacial prosthetic system. The material was formulated to have superior tear strength, low hardness and comparable viscosity in comparison to the main commercially used materials. Simple tensile tests were performed and a suitable constitutive formulation was proposed and validated in a commercial finite element code. This preliminary study will serve as a basis to introduce more complex features such as viscoelasticity and wrinkling of the soft polymeric structure in order to optimise the performances of the final prosthetic material.

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