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Filament-wound composite soft tissue prostheses:controlling compliance and strength by water absorption and degradation

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This paper deals with the effect of water absorption on the mechanical performance of selectively biodegradable filament-wound composite soft tissue prostheses. It shows how the water absorption phenomenon can be utilized as a new concept of material design, in contrast to the current view that associates moisture absorption by composite materials exclusively with damage. Harnessing water absorption to property design of vascular grafts has two advantages. The first is the controlled increase of the compliance of the graft during healing, aiming to reach a final stage of isocompliance with the host artery. The second is the water-induced biodegradation which enables regulation of the mural porosity during healing of an initially impervious graft. Filament-wound composite vascular prostheses comprising partially biodegradable matrices, different compositions and various winding angles are studied. Water absorption and material degradation are expressed in terms of relative weight gain/loss, which in turn is correlated with changes in the compliance and in the ultimate strength of the grafts.

1. Introduction

Filament-wound composite arterial prostheses have been introduced by a number of recent papers which describe the principles of the technology [1], the elastic properties [2] and the medical compliance and the ultimate strength of the prostheses [3]. The results of a 3-month implantation study in dogs of 6 mm diameter prostheses were also reported [4]. These papers emphasize two major features of the filament-wound arterial prostheses, namely, their winding angle controlled elastic and ultimate mechanical properties, and their partially biodegradable matrix.

It is widely accepted now that a major requirement for optimal healing and patency of a vascular graft is matching of its mechanical properties to those of the anastomosed natural vessels. This approach is advocated by what is known as the 'isocompliance concept'. The medical compliance of an artery, C_M, is defined as the relative internal volume change per an internal pressure change. It can be shown that $C_M = 2(\Delta R/R_0)/\Delta P + (\Delta L/L_0)/\Delta P$, where R_0 and L_0 are the internal radius and the length of the tube, ΔR and ΔL are radius and length changes and ΔP is the pressure change. Assuming that the anisotropic function of the artery is similar to that of an infinite open thin-walled tube, the internal pressure results in a pure hoop loading situation, with no loading or restriction of movement of the tube in the axial direction. Because the total length of the artery is constant, its overall response to an internal pressure change is often approximated to a cross-sectional area change at a constant length, i.e., $C_M = 2(\Delta R/R_0)/\Delta P$. For Hookean behaviour, the medical compliance is related to the engineering compliance, C_E, or to the Young's modulus, E, by: $C_M = (2R_0/t)C_E = (2R_0/t)/E$, where t is the wall thickness [3, 5]. It can be seen that as the engineering compliance is inherently a material property, the medical compliance depends on the geometry of the artery, and for a given material, the stiffness of the tube will increase if the diameter is decreased or the wall thickness is increased.

Lack of compliance matching with the host artery is detrimental to the acceptance of synthetic vascular grafts, when used in reconstruction of arteries. A compliance mismatch results in a mechanical incongruity, and in a blood flow of high shear stress and turbulence, with local stagnations. These factors may lead to local thrombosis, and may damage the arterial wall [5].

The use of biodegradable materials in arterial reconstructive surgery produces a graft which functions as a temporary scaffold so that the ingrowing tissue will eventually take over the mechanical function of the graft [6–9]. Porosity is regarded as an important

factor in graft healing [10]. An important advantage of using biodegradable materials is in their ability to seal the pores in the non-absorbable parts of the grafts at the time of implantation, preventing any loss of blood. The absorption of this material during the healing process will result in the ingrowth of granulation tissue which will provide a stable anchorage for the development of a viable cellular lining. The optimal pore size of the outer and inner layers of the graft can be designed to meet the exact needs of this ingrowth and anchorage.

The present paper deals with the effect of water absorption on the mechanical performance of selectively biodegradable filament-wound composite prostheses. It shows how mural hydration of the prosthesis can be integrated into the material design concept, in contrast to the current view that focuses on the damage which is associated with water absorption by composite materials. This study discusses two effects of water absorption, namely, the post-implantation controlled increase of the compliance of the prosthesis, and the hydrolytic biodegradation, which enables regulation of the porosity during healing. The experimental work is carried out on filament-wound composite vascular prostheses of different winding angles and different compositions.

2. Materials and experimental methods

Composite grafts were made of Lycra monofilament fibres in a matrix of a Pellethane/PELA mixture, where PELA denotes a block copolymer of lactic acid (LA) and polyethylene glycol. The details of the materials and of the filament winding procedure have been given previously [1–3]; however, a TiO_2 filled Lycra fibre (T-126) of 40 denier was used here instead of the clear fibre (T-336-C) used previously.

The geometrical data of the filament wound tubes were as follows: internal diameter 4 mm; wall thickness 0.30 mm; length 60 mm. The material composition varied across the wall thickness, whereby the luminal and central layers had a higher matrix weight fraction and were richer in the biodegradable component. The luminal and central layers (amounting to 2/3 of the wall thickness) were composed of PELA/Pellethane at an 80:20 weight ratio, and a total matrix weight fraction of 0.80. The outside 1/3 of the wall thickness had a PELA/Pellethane weight ratio of 50:50, and the total matrix weight fraction was 0.67.

Large diameter tubes (70 mm) were used to prepare tensile specimens as follows. After completing the winding process, the filament wound tube was cut axially (in parallel to the mandrel axis), spread out to form a flat rectangular laminate, and cut into 5 mm by 70 mm strips. Here, a uniform matrix composition of 70:30 PELA/Pellethane was maintained across the thickness, with a total matrix weight fraction of 0.75.

In this work, two parameters were varied and tested independently: the winding angle, θ, and the chemical composition of the biodegradable component. Three winding angles of 30, 50 and 70° with respect to the axis of the tube, and three PELA compositions (denoted PELA 1.2, 1.0 and 0.8) were tested. The nominal copolymer compositions comprised 0.005 mol of 3400 MW polethylene oxide (PEO) blocks per 1.2, 1.0 or 0.8 mol, respectively, of lactic acid.

Weight change experiments were carried out by immersing the samples in a Hartman solution at 37 °C. Sample weights were recorded at regular time intervals by removing samples from the solution bath, and drying with blotting paper prior to weighing. The dry weight of each sample was also determined after drying to a constant weight in an oven at 50 °C. Denoting the original, the wet and the dry weights M_o, M_w and M_d, respectively, a number of weight change parameters could be defined and calculated as follows:

$\Delta M_{w,o}$ the relative total weight change:

$$(M_w - M_o)/M_o$$

$\Delta M_{o,d}$ the relative weight loss, due to degradation:

$$(M_o - M_d)/M_o$$

$\Delta M_{w,d}$ the relative net moisture uptake:

$$(M_w - M_d)/M_d$$

Compliance testing was performed on wet specimens, as retrieved from the immersion bath, with the apparatus in Fig. 1, that enabled pure hoop stressing of the tubes under internal pressure conditions. Strains were calculated from diameter changes measured with a coordinate cathetometer (Gaertner), accurate at the 1μm level, attached to a digital readout processor (Quadra-Check II, Metronics, Inc.). The relative radial change, ΔR, was plotted against the pressure ΔP, and the compliance, defined as $C_M = 2(\Delta R/R_o)/\Delta P$, was taken as the slope of the initial part and was presented in 10^{-2} %/mmHg, the common units of medical compliance.

Tensile testing was performed on wet strip specimens by loading them at a gauge length of 50 mm and a loading rate of 5 mm min^{-1}, producing a nominal strain rate of 0.17% s^{-1}. The ultimate strength was determined from the maximum load at the point of failure, and the tensile modulus was calculated from the slope of the initial portion of the stress–strain curve.

3. Results and discussion

3.1. Mechanical behaviour

The exposure to Hartman solution is assumed to simulate physiological conditions under which hydration, degradation and mass loss of the matrix are expected to occur in that sequence. The result of this

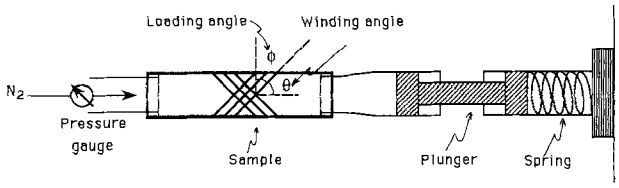


Figure 1 A schematic description of the apparatus used for quasi-static measurement of the hoop compliance of conduit prostheses under pure hoop stress state. ϕ is the loading angle with respect to the fibre direction.

three-stage process will be a change in the elastic and ultimate mechanical properties of the prosthesis. In addition, some of these processes may have separate effects on a specific property; for example, hydration will result in increasing compliance due to plasticization of the polymer matrix. Although both the degradation and the plasticization of the biodegradable matrix depend on water diffusion, the mechanism of water transport as such is beyond the scope of this article. The anisotropic moisture diffusion mechanisms in composite materials have already been established [11], showing that the diffusion coefficient in the direction of the fibre is significantly higher than that in the perpendicular direction. Nevertheless, the water transport through the hydrophilic wall of the prosthesis is substantially different. Here, the diffusion through the arterial wall is expected to be mostly uniaxial, perpendicularly to the fibre direction. Hence, the prime interest of this study is the mechanical properties and how they can be tailored through water uptake considerations.

Fig. 2 presents the results of the hoop compliance of the conduits as a function of the immersion period in Hartman solution. Results are given for three loading angles, ϕ, with respect to the hoop direction (see Fig. 1). The results show a large increase (about three-fold) taking place over a period of 15 days, and followed by a levelling off. Fig. 3 presents the corresponding modulus results, obtained by converting the medical compliance data of the 4 mm diameter, 300 μm wall thick tubes. The values exhibited by the $\phi = 50°$ material are smaller than those of the two other materials. This is consistent with the theory [2], which predicts a minimum for that angle in the value of the modulus as a function of the loading angle. It is noted again that whereas the modulus and the engineering compliance are material properties, the medical compliance depends on the geometry of the artery. Thus, a modulus of 5 MPa, for example, translates to a compliance of either 3.6 or 4.5 10^{-2} %/mmHg in in tubes of wall thickness 300 μm and a diameter of either 4 mm or 5 mm, respectively. A comparison with literature values of the modulus or of the medical compliance of natural arteries shows that while initially the filament wound prostheses are more rigid, the

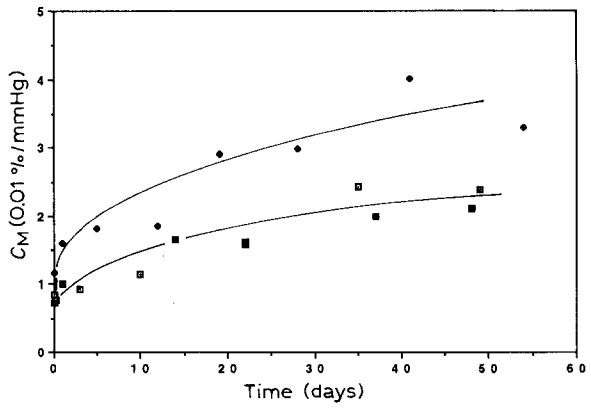


Figure 2 The hoop compliance of filament wound partially biodegradable tubular prostheses as a function of exposure period to physiological solution (■ $\phi = 20°$; ◆ $\phi = 40°$; □ $\phi = 60°$).

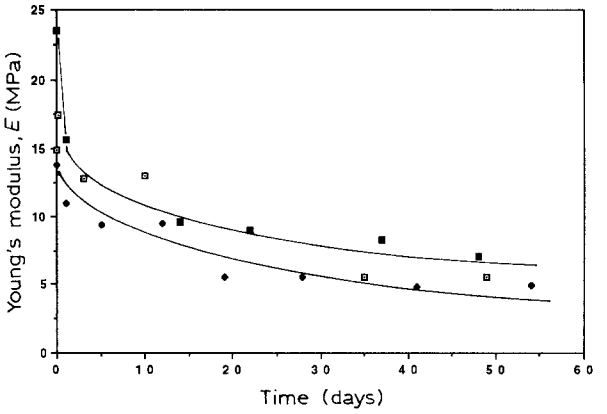


Figure 3 The engineering modulus values of 4 mm diameter, 300 μm wall thick tubes of the compliance values of Fig. 2 (■ $\phi = 20°$; ◆ $\phi = 40°$; □ $\phi = 60°$).

hydration process results in values which are comparable to, though still stiffer than, those of natural arteries. For example, compared with a modulus of 5 MPa of the hydrated filament wound prostheses, the tensile modulus of the canine left circumflex coronary artery is 0.77 MPa [12], that of a canine carotide artery is 0.88–1.95 MPa, under 80–140 mmHg [13] and those of the canine iliac artery at 100 mmHg transmural pressure are 1.3 and 0.4 MPa, in the hoop and axial direction, respectively [14]. Also, a compliance of 4.5 10^{-2} %/mmHg is low compared with that of 27.8 10^{-2} %/mmHg at 100 mmHg, reported for a canine carotide artery of 5 mm external diameter and 300 μm wall thickness [5].

A fuller picture of the effect of water on the modulus was achieved with the tensile specimens, where a wider range of loading angles, ϕ, was examined, as presented in Fig. 4. Although the modulus values are lower by a factor of 2 than those measured with the tubes (as attributed before to an 'edge effect' [3]), the overall trend is similar, showing a plateau in the hydration and degradation effects after about 15 days. Again, the lowest values are obtained for loading angles of 45° and 60°, in agreement with the theoretical analysis [2, 3].

Consideration of the ultimate mechanical properties of composite vascular prostheses is also very important because these structures are expected to operate under dynamic stresses, which may result in fatigue failure such as aneurysm. The ultimate strength of the filament-wound arterial prosthesis has been shown already to obey common failure criteria of composite materials [3]. This implies that the tensile strength is governed by either the fibre properties, by the matrix shear strength, or by the transverse strength of the composite, as the loading angle with respect to the fibre direction changes from 0° to 45° and to 90°, respectively. This obviously explains the results of the tensile testing presented in Fig. 5, showing the significantly higher values at $\phi = 0°$, dominated by the fibre properties, compared with the other angles, where the properties are matrix dominated. In general, the water-induced degradation produces some three-fold strength decrease after 10 days in Hartman solution. For comparison, the hoop stress

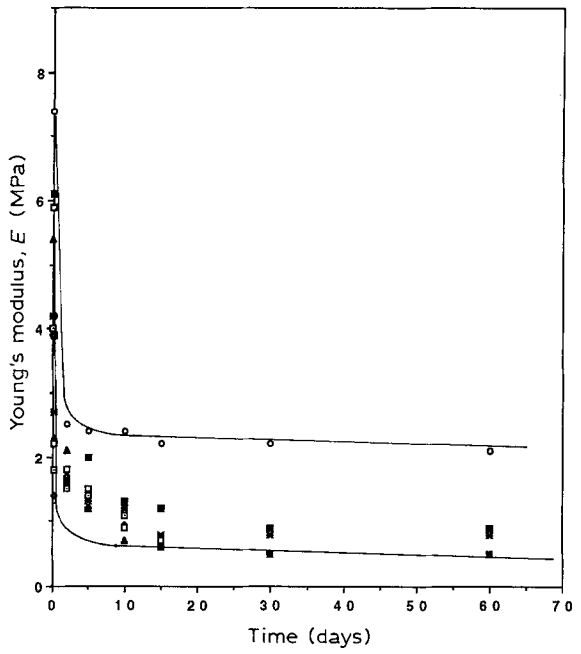


Figure 4 The effect of exposure to physiological solution on the modulus of partially biodegradable composite tensile specimens with a range of loading angles with respect to the reinforcement direction. (○ $\phi = 0°$; ■ $\phi = 20°$; ⋈ $\phi = 30°$; ◇ $\phi = 45°$; □ $\phi = 60°$; □ $\phi = 70°$; ▲ $\phi = 90°$)

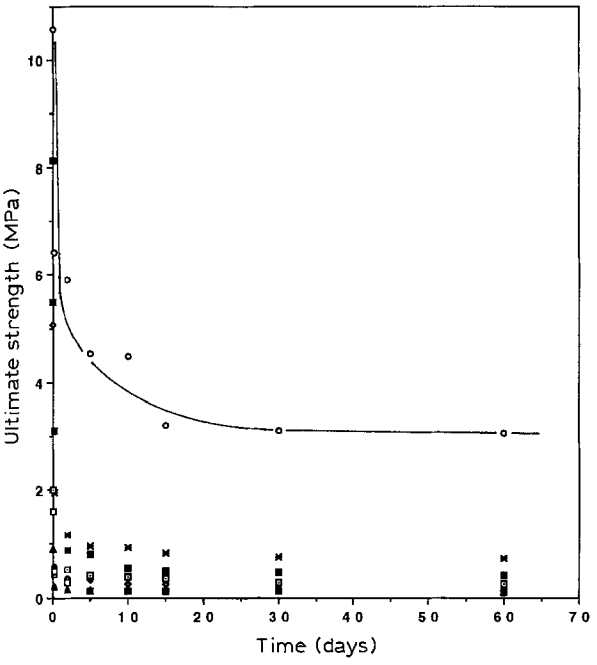


Figure 5 The ultimate tensile strength at different loading angles of partially biodegradable composites, as a function of the exposure period to physiological solution. (○ $\phi = 0°$; ■ $\phi = 20°$; ⋈ $\phi = 30°$; ◇ $\phi = 45°$; □ $\phi = 60°$; □ $\phi = 70°$; ▲ $\phi = 90°$).

which develops in the canine iliac artery of wall thickness 300 μm, under a pressure of 200 mmHg is 270 kPa [14], while the ultimate strength of the wet 30° prosthesis reaches after 15 days a plateau of approximately 1 MPa.

3.2. Water absorption

As pointed out, two effects may be responsible for the observed changes in the mechanical properties. These are the plasticization and the degradation of the matrix. By monitoring different values of weight change, as defined in section 2, the relative influence of each effect was determined.

The water diffusion behaviour during short exposure periods was apparently Fickian, as seen by the example in Fig. 6 for a $\phi = 60°$ sample. In this example, the total weight change is plotted against the square root of exposure time. It is seen that the substantial portion of the weight change process occurs within 1.5 h of exposure, when some 50% weight increase is reached. Based on such diffusion plots, the respective average coefficients of diffusion for the composites of PELA/Pellethane and of neat Pellethane matrices were 9.1×10^{-12} and 3.4×10^{-12} $m^2 s^{-1}$ (these values did not consider degradation and fibre orientation effects).

The results of the weight change as a function of exposure time to Hartman solution are shown in Fig. 7a–c. The results are given for PELA 1.2 based composite grafts of three winding angles relative to the hoop direction: 60, 40 and 20°. As the winding angle does not affect the nominal composition of the tubes, the observed differences between the three materials are unexpected. These differences could be attributed to inevitable fibre/matrix ratio variations associated with required technical changes in the filament winding process at different winding angles. The results show (Fig. 7a) that $\Delta M_{w,o}$ the relative total weight change that expresses the combined effect of weight gain due to water absorption and of weight loss due to degradation, goes through a maximum after approximately 15 days. This marks the point where the resorption of the biodegradable component becomes dominant, as confirmed by the observations that while the relative net water uptake $\Delta M_{w,d}$ stabilizes after 15 days (Fig. 7b), the relative residual weight due to degradation $1 - \Delta M_{o,d}$ continues to decrease monotonously (Fig. 7c). For comparison, some weight change results of the strip specimens are presented in Fig. 8a, b. Again, the unexpected differences observed for different angles are attributed to composition variations, as seen from the fact that specimen pairs produced from the same filament-wound sheets

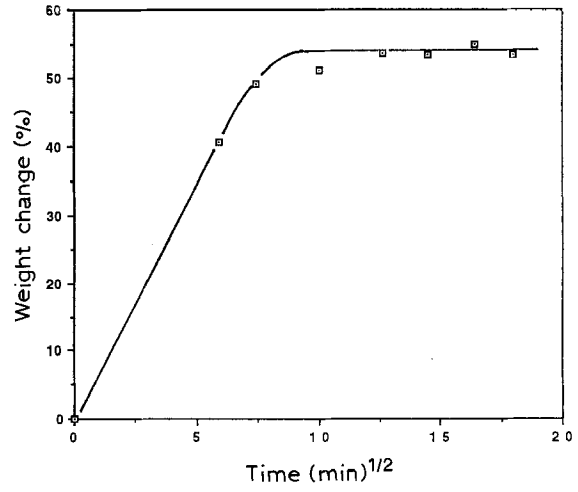
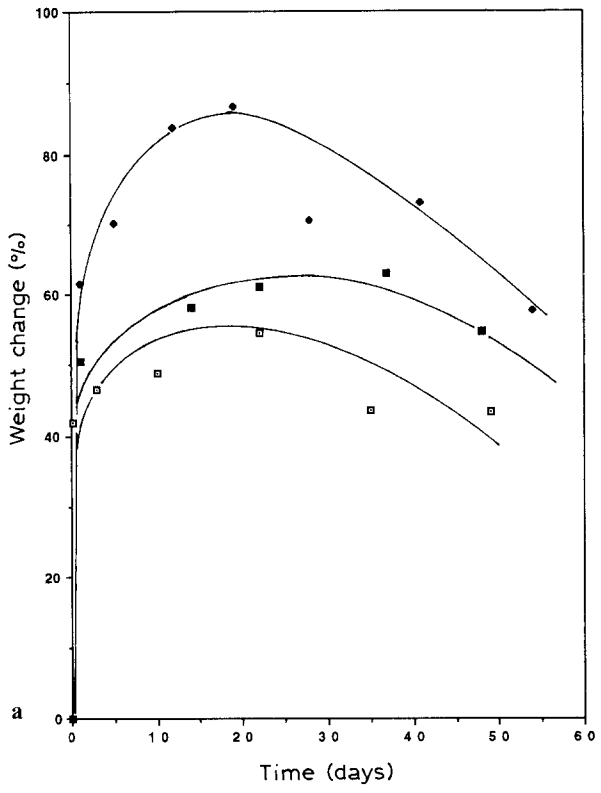


Figure 6 A plot of $\Delta M_{w,o}$, the relative total weight change, against the square root of exposure time for a $\phi = 60°$ sample, showing a nominal Fickian behaviour during the initial stage of the process.



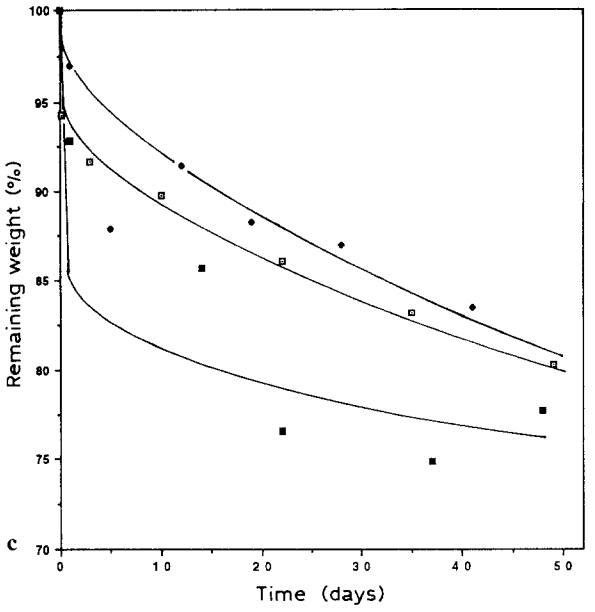
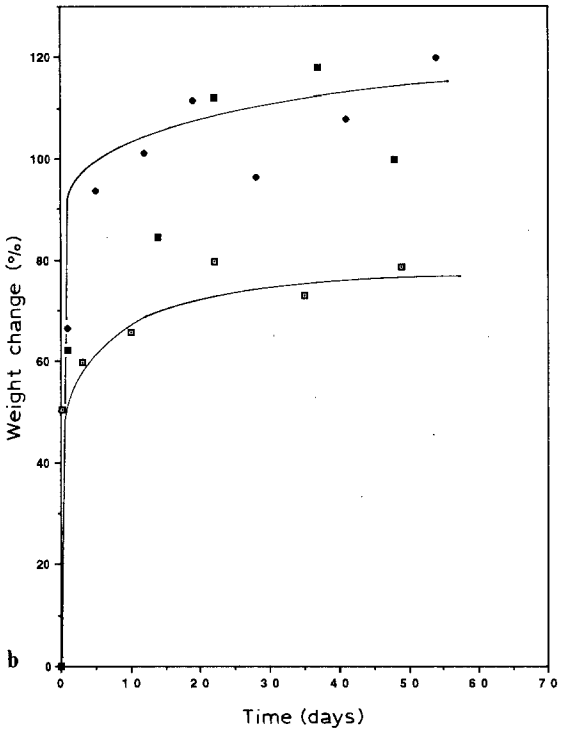


Figure 7 Relative weight change results of filament wound conduit prostheses as a function of exposure time to physiological solution: (a) $\Delta M_{w,o}$ the relative total weight change; (b) $\Delta M_{w,d}$ the relative net moisture uptake; (c)1 $-$ $\Delta M_{o,d}$ the relative residual weight (■ $\phi = 20°$; ◆ $\phi = 40°$; □ $\phi = 60°$).



(namely 0/90, 20/70 and 30/60) behave identically. Whereas the values of the relative total weight change $\Delta M_{w,o}$ are higher compared with those of the tubes (Fig. 7a), the remaining weight due to degradation $1 - \Delta M_{o,d}$ exhibits in the strip specimens (Fig. 8b) a similar behaviour to that of the tubes (Fig. 7c).

The weight change behaviour depends on the type and relative content of the biodegradable component, PELA. For example, Fig. 9 presents the relative total weight change for different PELA compositions compared with that of a pure Pellethane matrix tube. It is seen that the hydrophilicity of PELA increases with the ratio of PEO/LA in the block copolymer, which in turn increases the exposure of the esteric bonds to hydrolytic cleavage [15]. It is also seen that the Pellethane-based tube exhibits a very low water absorption capacity.

It is difficult to determine unequivocally which of the interrelated factors, the water absorption or the degradation, is responsible for the compliance increase and strength decrease, as these processes occur in part sequentially and in part simultaneously. Moreover, a linear ratio is maintained between their respective effects, on the one hand, while the compliance is correlated linearly with the weight loss, on the other, as demonstrated in Table I. However, a comparison of the property change data to these of the weight change may help establish the order of the main phases of the water interaction process and of their corresponding effects on the strength and compliance. Fig. 6 shows that most of the water uptake due to diffusion occurs within the first 2 h of exposure. At that stage of the process, the ultimate strength and modulus drop by about 50% due to plasticization of the matrix. Whereas this stage is very rapid, water-induced degradation is much slower, and takes about 20 days to level off. During that stage the strength and modulus continue to drop, mostly due to matrix degradation, while the continuing weight increase trend is accounted for by water molecules that fill the voids left by the resorbed polymer.

In vitro studies of the morphological manifestation of the degradation process have shown that transmural pores are formed in the prosthesis wall [16]. These, as shown by studies in animals [4], are then penetrated by a growing granulation tissue. Here, the pore formation as a result of the degradation process is exemplified in Fig. 10, by comparing scanning electron micrographs of an as-received sample with another, exposed to Hartman solution for 5 days.

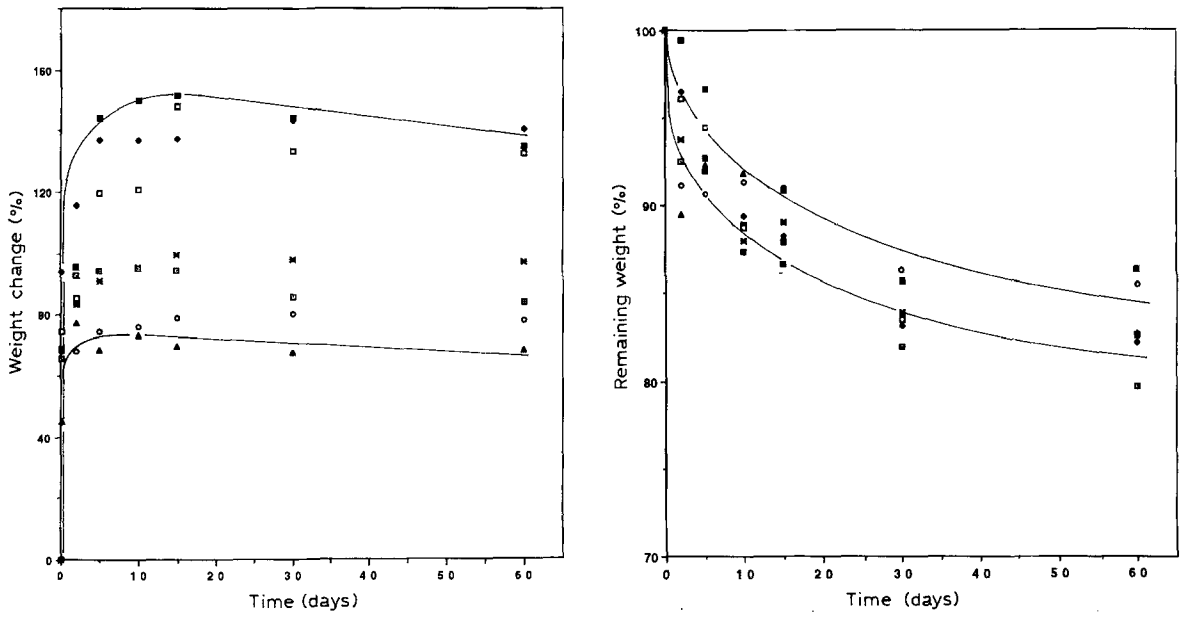


Figure 8 Relative weight change results of strip composite specimens as a function of exposure time to physiological solution: (a) $\Delta M_{w,o}$ the relative total weight change; (b) $1 - \Delta M_{o,d}$ the relative residual weight (○ $\phi = 0°$; ■ $\phi = 20°$; ⋈ $\phi = 30°$; ◇ $\phi = 45°$; □ $\phi = 60°$; □ $\phi = 70°$; ▲ $\phi = 90°$).

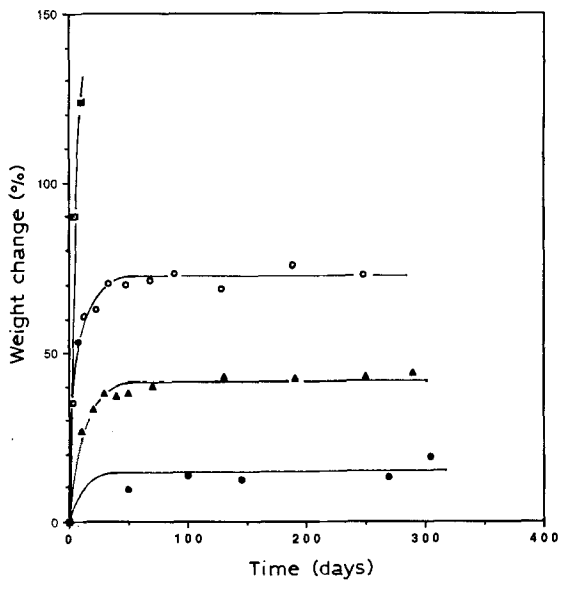


Figure 9 $\Delta M_{w,o}$ the relative total weight change results of filament-wound conduit prostheses as a function of exposure time to physiological solution: the behaviour of different PELA compositions compared with that of a pure Pellethane matrix (□ PELA 0.8; ○ PELA 1.0; ▲ PELA 1.2; ● Pellethane).

4. Conclusions

The main incentive for using biodegradable polymers in synthetic implants is to allow, or often even to trigger, a process which will ultimately result in a certain extent of replacement of the synthetic material by natural tissue. The changes in the mechanical properties of the implant due to its selective biodegradation and resorption must be accounted for, because the mechanical properties comprise an essential performance consideration.

Considering the partially biodegradable filament-wound vascular prostheses, the present *in vitro* simulation of the degradation process shows that the hydration and polymer resorption processes produce an increase in the compliance and a decrease in the strength of the prosthesis. It is possible to tailor the effect of these phenomena into the design of the implant, so that the desired mechanical properties are progressively achieved, It was shown, for example, that when the biodegradable component consisted of PELA 1.2, a modulus which is of the same order as, and a strength which is an order of magnitude higher than that of natural arteries, are achieved.

TABLE 1 Compliance increase versus weight changes due to hydration and mass loss

Loading angle								
$\phi = 20°$			$\phi = 40°$			$\phi = 60°$		
C_M	$\Delta M_{w,d}$	$\Delta M_{o,d}$	C_M	$\Delta M_{w,d}$	$\Delta M_{o,d}$	C_M	$\Delta M_{w,d}$	$\Delta M_{o,d}$
(0.01 %/mmHg)		(%)	(0.01 %/mmHg)		(%)	(0.01 %/mmHg)		(%)
0.72	57.7	7.2	1.17	64.7	3.0	0.84	47.5	5.7
1.01	72.4	14.3	1.59	82.4	12.1	0.78	54.7	8.3
1.65	84.6	23.4	1.82	92.5	8.5	0.92	59.0	10.2
1.62	88.2	25.1	1.86	98.6	11.7	1.15	68.3	13.9
1.99	77.0	22.3	2.91	83.8	13.0	1.59	60.4	16.8
–	–	–	2.98	85.8	16.5	2.42	63.0	19.7

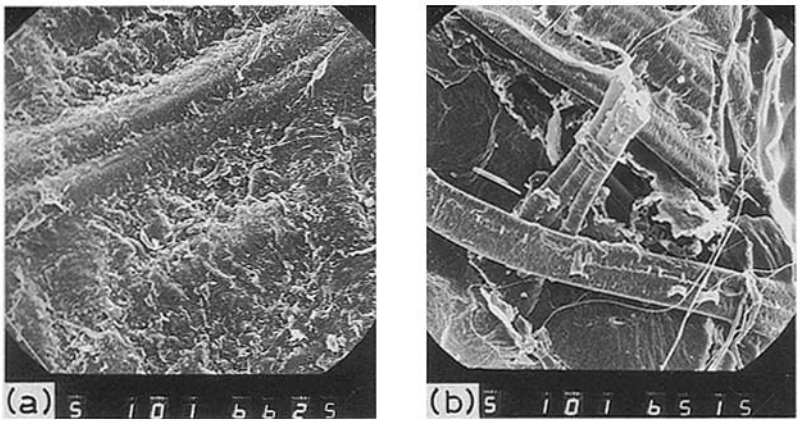


Figure 10 Scanning electron micrographs of the external mural surface of an as received filament wound conduit (× 50) after ethylene oxide sterilization (a) and of the same sample after a 5-day exposure to physiological solution (b).

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