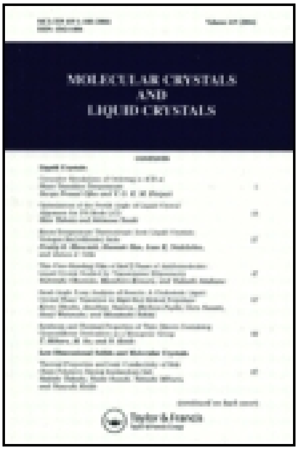

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Zelfo – An Engineering Material Fully Based on Renewable Resources

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The development of *Zelfo* was based on a review on technologies for the production of "papier mâche" in the second half of the 19[th] century. However, in contrast to "papier mâche" *Zelfo* does not contain any additional bonding agent. Potential raw materials for the production of *Zelfo* include various plants and wastes with a high cellulose content (e.g., hemp, flax, waste paper). The first production step is a refining process, in which the raw materials are simultaneously hackled and ground and mixed with water. The result of this process is a microfiberpulp with a solid content of 1 to 15 m%, which is subsequently predried, cast or molded into a final shape and dried. Due to the possible variations in raw materials and in the production process, properties of *Zelfo* can be varied over a certain range. For example, values for density from 0.5 to 1.5 g/cm^3, for tensile modulus from 1500 to 6550 MPa and for tensile strength from 7 to 55 MPa can be achieved. Thus, *Zelfo* is likely to compete with both conventional plastics and chipboard for certain applications.

Keywords: zelfo; material; cellulose; renewable resources; biodegradable

INTRODUCTION

Starting point for the development of *Zelfo* was a review study on materials based on renewable resources and corresponding processing technologies, particularly of "papier mâche", which gained importance in the second half of the 19[th] century. At that time "papier mâche" was used for a wide variety of applications including furniture, building and construction products, household appliances, music instruments, etc. At the beginning of the 20[th] century materials based on "papier mâche" were gradually replaced by Bakelite® and other plastics.

While the production of "papier mâche" also included the addition of separate bonding agents, it was the aim of the development of *Zelfo* to obtain cellulose based materials without any additional bonding agent. Hence, extensive practical experiments were carried out to enhance the intrinsic bonding capability of the cellulose fibers. As a result, materials with reasonable mechanical properties fully based on renewable resources could be produced.

The purpose of this paper is to provide an overview of the production process and the range of basic mechanical properties of the *Zelfo* material class. The *Zelfo* property profiles are compared with those of typical commodity plastics and wood containing materials.

RAW MATERIALS AND PROCESSING

The range of potential raw materials for *Zelfo* includes a vast number of plants with high cellulose content such as hemp, flax, straw, reed, sugar cane, sisal and alike, as well as cellulose containing consumer and industrial waste products (e.g., old textiles and waste paper). The plant based renewable raw materials can be used directly as complete plants or as special fractions of various other plant preparation processes (e.g., hemp scrape, straw dust). All these raw materials can be used in pure form or in mixtures of various compositions.

A scheme of the production process of *Zelfo* is shown in Fig. 1. In a first step, the raw materials are simultaneously compounded with water and hackled in a refiner yielding the so-called microfiberpulp as intermediate product. To obtain the desired microfiberpulp consistency, a specific grinding energy is required. The microfiberpulp with a solid content of 1 to 15 m% is then exposed to a pre-drying process, which can take place with or without pressure.

The subsequent casting or molding process may be carried out in several steps. Each of these processing steps is usually followed by an additional interim drying stage whenever shapes with complicated geometry are to be pro-

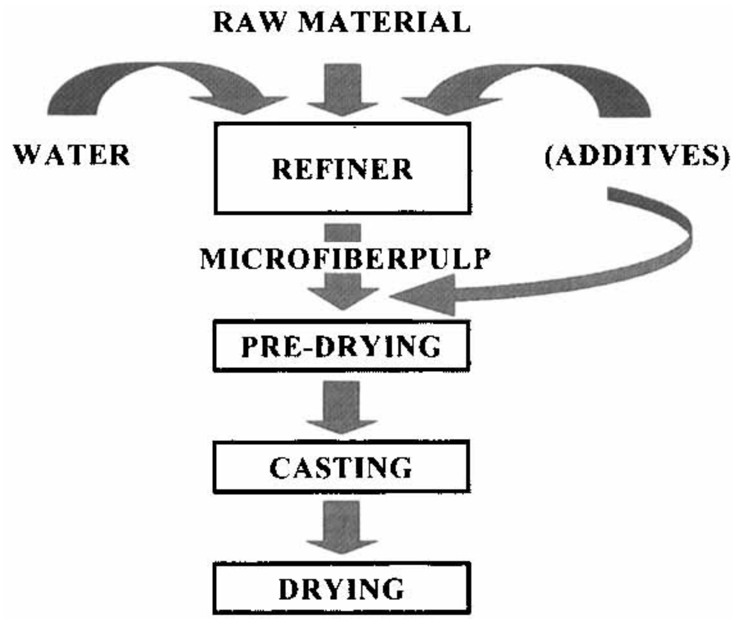


FIGURE 1 Production process scheme of *Zelfo*

duced. The casting and molding steps can take place without and with pressure, respectively. As *Zelfo* materials absorb relatively high amounts of moisture which substantially enhances the material flexibility, they may be reshaped to increase the dimensional precision of parts or components upon additional wetting and post-forming procedures. The wetting process can be carried out either by storing dry sheets or products in a steam saturated climate chamber for several hours up to days (depending on the maximum wall thickness and the necessary deformation) or by immersing these products in water. Subsequent drying hardens *Zelfo* again to previous values of density, stiffness and strength.

In general, moldability, density, stiffness and strength were found to increase with growing refinement of the cellulose containing raw material. However, best mechanical properties in terms of high stiffness and strength values can be achieved by addition of a certain amount of less refined longer cellulose fibers to the finely ground microfiberpulp. While the finely ground microfiberpulp in these mixtures apparently acts as a bonding agent with higher density values reflecting a higher degree of compaction, the coarse cellulose fibers are believed to act as second phase reinforcement similar to short fibers in short fiber reinforced plastics.

Depending on the application it may also be useful to add further additives before the refining process or to the microfiberpulp. Thus, *Zelfo* products with higher fire resistance, better insulation properties, different colors, etc. are obtained.

To achieve porous materials of lower density for light weight parts and components, two possibilities exist. In one process route the microfiberpulp is frozen after the shaping process and subsequently dried. In the second process route propellants or ferment agents are added to the microfiberpulp to achieve a microstructure of a certain porosity.

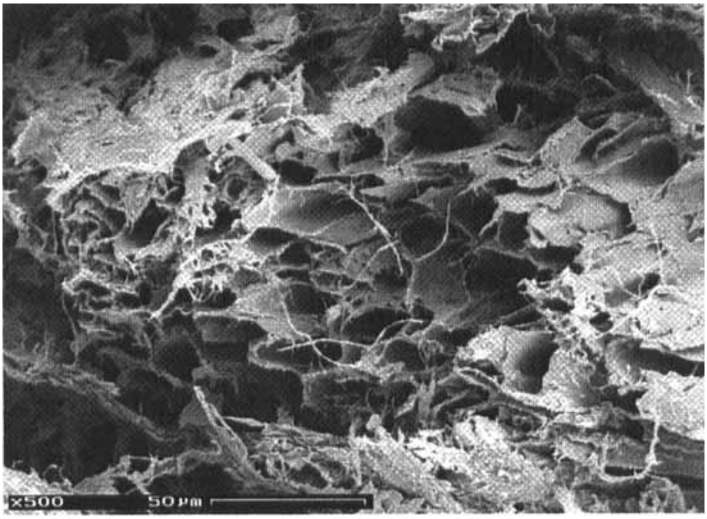
MATERIALS, MATERIAL MICROSTRUCTURE AND PROPERTIES

Materials

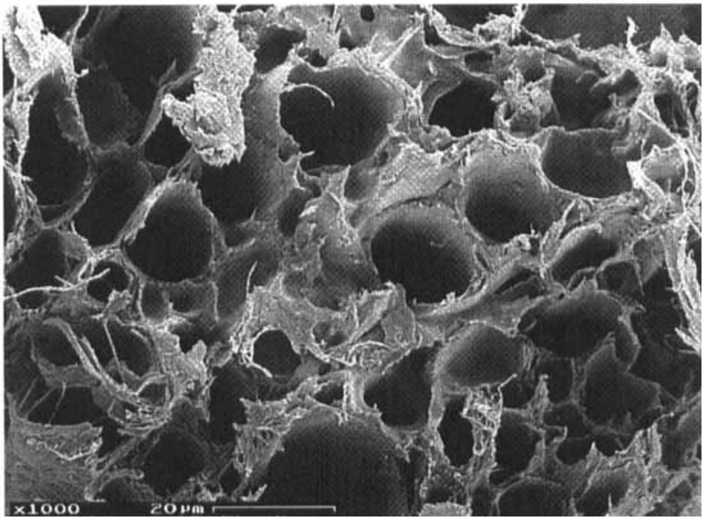
For the purpose of the present paper two material types of the *Zelfo* material class were investigated. The one material, designated *Zelfo* HG, was produced directly from hemp plants, the other material, designated *Zelfo* HZ, from refined hemp cellulose. Processing procedures and parameters for both materials were selected to achieve optimum mechanical properties.

Material Microstructure

The material microstructure as observed on typical fracture surfaces is shown for *Zelfo* HG and *Zelfo* HZ in Fig. 2 and Fig. 3, respectively. These micrographs were obtained by scanning electron microscopy (SEM) using a Zeiss SEM device of the type DSM 962 with an LaB_6-cathode (Zeiss, Oberkochen, D). While the fracture surface of the lower density material



(a)



(b)

FIGURE 2 Fracture surface appearance and material microstructure of *Zelfo* HG

FIGURE 3 Fracture surface appearance and material microstructure of *Zelfo* HZ

Zelfo HG in Fig. 2a and 2b reflects a coarse microstructure with certain a degree of porosity associated with the use of unrefined hemp, the microstructure of the higher density material *Zelfo* HZ in Fig. 3 reveals a higher degree of compaction with more finely dispersed fibrils mainly due to the additional refining process of the hemp cellulose raw material.

Material Properties

To characterize the material properties of the two *Zelfo* types, appropriate specimens were machined from *Zelfo* sheets of approximately 4 mm thickness. With the exception of the Charpy impact specimens for *Zelfo* HZ (which were obtained from original sheets without surface grinding), all other specimens were obtained from compression molded sheets with ground surfaces to reflect the surface condition of typical commercial *Zelfo* products.

The results of the various investigations performed on both *Zelfo* materials are listed in Table 1. All specimens were stored at 23 °C and 50 % r.h. for at least one week prior to testing. For comparison also provided in Table 1 are typical properties of other materials such as the commodity plastics high-density poly(ethlyene) (PE-HD) and unplasticized poly(vinylchloride) (PVC), and wood meal filled phenolic resin (PF-HM) and chipboard. The data for the latter materials were obtained from the appropriate literature [1-3]. The properties listed in Table 1 will be discussed in more detail below.

TABLE 1 Comparison of properties of *Zelfo* materials with commodity plastics (PE-HD, PVC), wood meal filled phenolic resin (PF-HM) and chipboard [1-3]

	Density	*Tensile*		*Flexural*		*Charpy Impact*	
		Modulus	Strength	Modulus	Strength	Strength [kJ/m²]	
	[g/cm³]	[MPa]	[MPa]	[MPa]	[MPa]	unnotched	notched
Zelfo HG	0,5	1500	7	1800	17	2	1
Zelfo HZ	1,5	6550	55	9400	95	14[a)]	6[a)]
PE-HD	0,95	1000	25[b)]	-	-	-[c)]	15
PVC	1,4	3000	60[b)]	-	100	-[c)]	≥ 2
PF-HM	1,4	-	25	7000	≥ 70	≥ 6	≥ 1,5
Chipboard	0,7	-	-	1800	18	-	-

[a)] Unground specimens [b)] Yield strength [c)] No fracture

Density

The density range covered by *Zelfo* materials starts with *Zelfo* HG at 0.5 g/cm³ at the lower end reaching up to 1.5 g/cm³ for *Zelfo* HZ at the upper end (it should also be mentioned that density values as low as 0.3 g/cm³ may be obtained for porous *Zelfo* types as discussed above; such materials, however, were not part of the present investigation). For comparison, at the lower end of the *Zelfo* density range, chipboard reveals a density of approximately 0.7 g/cm³, while PVC and PF-HM show density values of 1.4 g/cm³, close to the upper end of the range of *Zelfo* materials. PE-HD, with 0.95 g/cm³, shows a density in the mid-range of densities covered by *Zelfo* materials.

Tensile properties

The tensile tests were carried out with an Instron 4505 universal electromechanical testing machine (Instron Ltd., High Wycombe, UK) according to ISO 527. Tensile modulus values of *Zelfo* HG and *Zelfo* HZ with 1500 MPa and 6550 MPa, respectively, differ by a factor of 4, reflecting the differences in density. These values compare very favorably with those of the other materials (PE-HD: 1000 MPa; PVC: 3000 MPa).

In terms of tensile strength, the differences between *Zelfo* HG and *Zelfo* HZ are significantly enhanced, revealing a difference of a factor of 8 (*Zelfo* HG: 7 MPa; *Zelfo* HZ: 55 MPa). Nevertheless, the tensile strength values for *Zelfo* materials are also comparable to those of the other materials, at least for the higher density range (PE-HD: 25 MPa; PVC: 60 MPa; PF-HM: 25 MPa). At this point it should also be mentioned that specimens of *Zelfo* with unground surfaces reveal significantly higher tensile strength values, with a tensile strength for *Zelfo* HZ of 75 MPa.

Flexural properties

Flexural tests according to ISO 178 were also carried out with an Instron 4505 testing machine. Due to non-linear elastic behavior and possible effects associated with a somewhat higher density in the surface skin layers of the compression molded specimens, both flexural modulus and flexural strength of *Zelfo* were found to be higher than the corresponding tensile properties. The flexural modulus values of 1800 MPa and 9400 MPa and the flexural strength values of 17 MPa and 95 MPa for *Zelfo* HG and *Zelfo* HZ, respectively, again compare well with the other materials listed in Table 1.

Impact properties

Impact tests were carried out with a Ceast pendulum of the type Resil 25 (Ceast Spa., Turin, I) following ISO 179 procedures. While for *Zelfo* HG an unnotched impact strength of 2 kJ/m^2 was found, *Zelfo* HZ reveals a value of about 14 kJ/m^2, however, with unground specimens in the latter case. For notched specimens these values are reduced to 1 kJ/m^2 for *Zelfo* HG and to 6 kJ/m^2 for *Zelfo* HZ, again using unground specimens for *Zelfo* HZ. While this data cover the range of PVCU, PF-HM and probably also chipboard, for which no data were available, ductile plastics such as PE-HD and other highly ductile plastics reveal substantially improved impact behavior.

Effect of water immersion on flexural properties

To study the effects of water immersion on flexural properties on *Zelfo* HZ, specimens were immersed in distilled water at 23 °C for time periods from 1 to 168 h according to ISO 175. Subsequently one set of test specimens, designated "Wet", was investigated in flexural tests immediately after the water exposure, while another set of test specimens, designated "Re-conditioned", was first re-conditioned at 23 °C/50 % r.h. to constant weight and then tested for flexural properties. To study the reproducibility of the sheet production process, specimens from two different sheets were included for each of these test series. The flexural tests were again carried out following ISO 178 procedure using an Instron 4505 testing machine.

The effects of these specimen conditioning procedures on the flexural properties of *Zelfo* HZ are shown in Fig. 4. From these results it is clearly apparent that a drop-off in both modulus and strength by a factor of approximately 20 occurs within the first 24 h of water immersion for wet specimens. From 24 h to 168 h of water immersion no further significant changes in properties were observed. While the significant reduction in properties during the first 24 h of water immersion may be not tolerable from an application point of view, so that intensive water exposure of these *Zelfo* types during use should be avoided, it is of course advantageous from the standpoint of reshaping and reprocessing of these materials.

In this context of particular importance are the findings, that both modulus and strength increase to original values once the water content of the material re-equilibrates to values prior to water immersion (compare data for re-conditioned materials in Fig. 4). In other words, from these results it may be

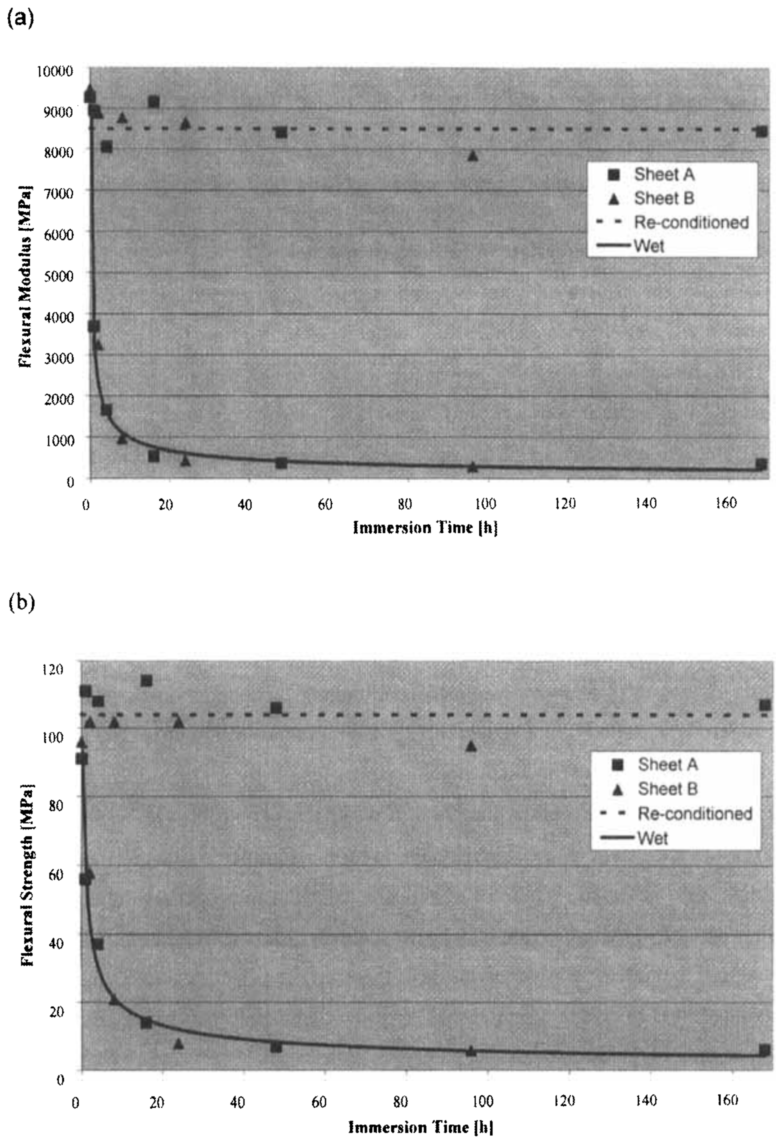


FIGURE 4 Effect of water immersion and re-conditioning of *Zelfo* HZ on (a) flexural modulus and (b) flexural strength

concluded that the effect of water uptake during immersion is of pure physical nature without any significant chemical degradation.

Finally, the comparison of data obtained from specimens of two different sheets indicates a high degree of reproducibility, at least for the compression molding process. Further studies will also investigate reproducibility effects associated with different raw material batches and production processes.

SUMMARY AND CONCLUSIONS

The purpose of this paper was to provide an overview of the production process and to describe the mechanical behavior of *Zelfo* materials, which are fully based on renewable resources with high cellulose content (hemp, flax, straw, waste paper, etc.). As no other non-renewable substances are required, one of the big advantages is related to the biodegradability of these materials.

The preliminary investigations performed reveal that the mechanical property profile of *Zelfo* materials compares well with mechanical properties of other materials such as commodity plastics (PE-HD, PVC), PF-HM and chipboard. Due to the cellulose based chemical nature of *Zelfo*, *Zelfo* materials readily absorb water which significantly reduces modulus and strength properties. However, water immersion and re-conditioning experiments (23 °C/50 % r.h.) indicate that the effect of water uptake is of pure physical nature without any significant chemical degradation.

Future investigations will focus on additional mechanical properties including the time/loading rate effects and the influence of test temperature as well as on obtaining a better understanding of structure-property-relationships using fracture mechanics methods.

Acknowledgments

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