TRANSVERSE PERMEABILITY AND KOZENY CONSTANT IN FLAX FIBER MATS PREFORMS

Laurent Bizet [1, 4], Pierre Ouagne [1], Joël Bréard [1], Christophe Baley [2], Jean-Paul Jernot [3] and Moussa Gomina [3]

[1] *Laboratoire d'Ondes et de Milieux Complexes (LOMC), Université du Havre,*
53 rue de Prony, F-76600 Le Havre, France
[2] *Laboratoire d'Ingénierie des MATériaux de Bretagne LIMATB, Université de Bretagne Sud,*
rue de saint Maudé, BP 92116, F-56321 Lorient cedex, France
[3] *ESCTM du CRISMAT, UMR 6508 ENSICAEN / CNRS, 6 Boulevard Maréchal Juin,*
F-14050 Caen cedex, France, Email: jean-paul.jernot@ensicaen.fr
[4] *Corresponding author's Email: laurent.bizet@univ-lehavre.fr*

SUMMARY: New composite materials based on flax fibers and biopolymer matrixes are currently under development. Impregnation of flax fibers must be studied in order to improve the manufacturing of such materials. The present work deals with transverse permeability of composite preforms made of flax fibers mats. Saturated permeability has been measured with an original experimental device. Permeability values are well within the range that allows an easy transverse impregnation. A process like "Film Stacking" can then easily be used to manufacture composite parts with a flax mats preform. An interpretation of the permeability values on the basis of Kozeny-Carman equation shows that the permeability may be improved by modifying the hackling of fibers.

KEYWORDS: flax fibers, mats preforms, transverse permeability, Kozeny constant

INTRODUCTION

A new type of composite materials is currently under development. They are called either biocomposites, ecocomposites or green composites and are devoted to the limitation of the negative environmental consequences of human activities. Two major impacts of production and use of materials on ecosphere are resources consumption and waste disposal. Comparisons between materials according to their composition, elaboration, use or disposal can be made but difficulties remain to estimate the overall benefit of choosing a material instead of another one. Consequences may occur, for example, either on air, water or earth parts of the ecosphere. The composite materials are optimum for a specific use: they are generally lightweight and minimize energy consumption, e.g. when used in transportation's systems. But their global environmental impact, including manufacturing and end-of-life, must also be considered. For example, it has

been shown that natural fibers used in composite materials have qualitatively less environmental impacts than glass fibers [1-2]. Moreover natural fibers (such as hemp or flax) and glass fibers have similar specific mechanical properties [3]. Nowadays two types of studies have to be carried out for the development of ecocomposites: more environmental "friendly" matrices and new manufacturing processes. This work contributes to the latter subject.

In a previous study [3], a process called "Film Stacking" had been proposed to elaborate composite parts with flax fibers and a biodegradable thermoplastic polymer. In this process, the flax fibers are assembled into mats – scattered onto a plane surface– and the polymer is shaped into films. Then the flax mats and the polymer films are stacked sequentially. Finally, the whole preform is heated and compressed such as the biopolymer melts and flows through the mats.

Firstly the "Film Stacking" process is preferred because the viscosity of the molten biopolymer is high: typical viscosity values range from 100 Pa.s for the thinnest ones to more than 1000 Pa.s [3] for polymers with best mechanical properties.

The second reason to choose the "Film Stacking" process for this type of material comes from the difficulty to impregnate a preform elaborated with flax fiber mats. As a matter of fact, a previous result shows that longitudinal permeability of flax mats is about three times lower than that of glass mats containing the same volume fraction of fibers [4]. "Film Stacking" process is characterized by a transverse flow through each mat of the preform. The distance the fluid flows is then significantly reduced. Therefore the "Film Stacking" process is preferred because the impregnation step is shortened during the manufacturing process cycle of the material.

This work presents experimental results and an interpretation of the transverse permeability as a function of the volume fraction of fibers in flax mats. Permeability measurements are made with a "model fluid" which flows through the entire preform. The interpretation of the results is based on the Kozeny-Carman relation [5]. The motivation of this work was to acquire knowledge on the "Film Stacking" process applied to composite materials made with flax fibers and biopolymers. The global goal here is to study the manufacturing processes of flax fiber composites.

FLAX FIBER MATS CHARACTERISTICS

Technical flax fibers (i.e. bundles of fibers) from Hermes variety are extracted from the plant by dew-retting and hackling after which they are cut into segments of about 10 mm length. These segments are agglomerated into flax mats by a paper manufacturing process. Flax mats can be easily handled and stacked for the elaboration of preforms. Fig. 1 shows the appearance of a flax mat (left) and the randomly scattered arrangement of the flax fibers (right).

Areal Weight of Flax Mats

Flax mats are partially characterized by their areal weight i.e. the weight of flax fibers per unit area of mat ($g.m^{-2}$). Areal weight of flax mats has been checked when samples were cut for the subsequent permeability experiments. One hundred sixty circular samples with diameter of about 10 cm - part of one sample is shown on the left of Fig. 1 - have been weighed. The average areal

weight is 116 g.m^{-2} (Fig. 2). This value is close to the 110 g.m^{-2} value defined before mats elaboration.

The areal weight of flax mat samples shows a relative standard deviation (6.7%) lower than that of an industrial glass mat (11.9%) on which the measurements have been made by following the same procedure as flax mats out of one hundred and fourteen samples. A comparison of areal weight distributions between flax and glass mats is reported in Fig. 2. The flax mat elaborated in our laboratory shows a better spatial arrangement of fibers than the industrial glass mat and should allow reproducible permeability measurements.
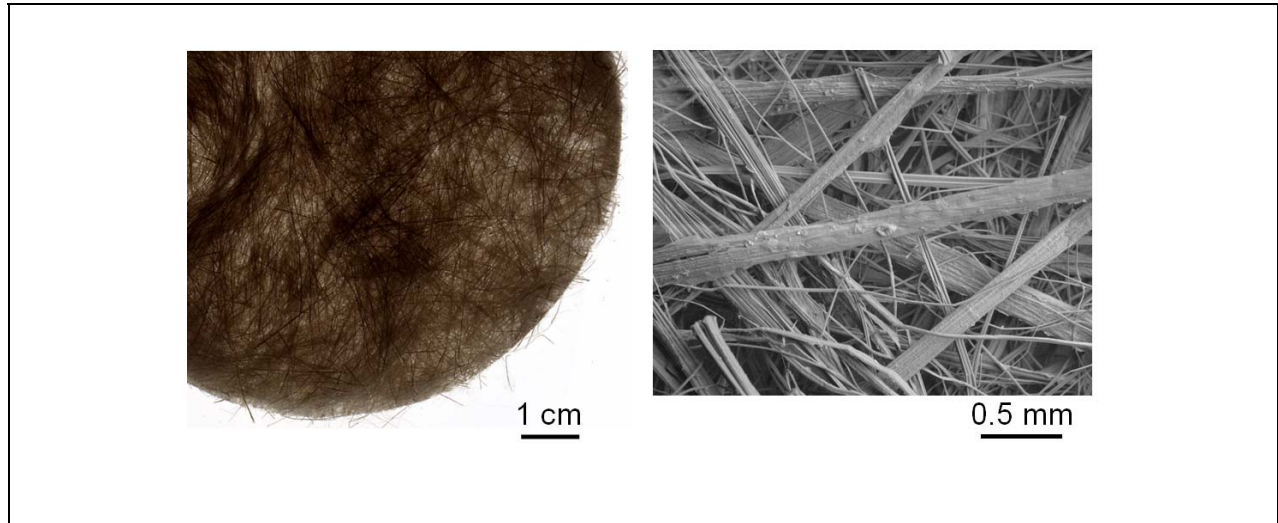

Fig. 1 Surface aspects of flax mat examined with an optical scanner (left) and with a scanning electron microscope (right).
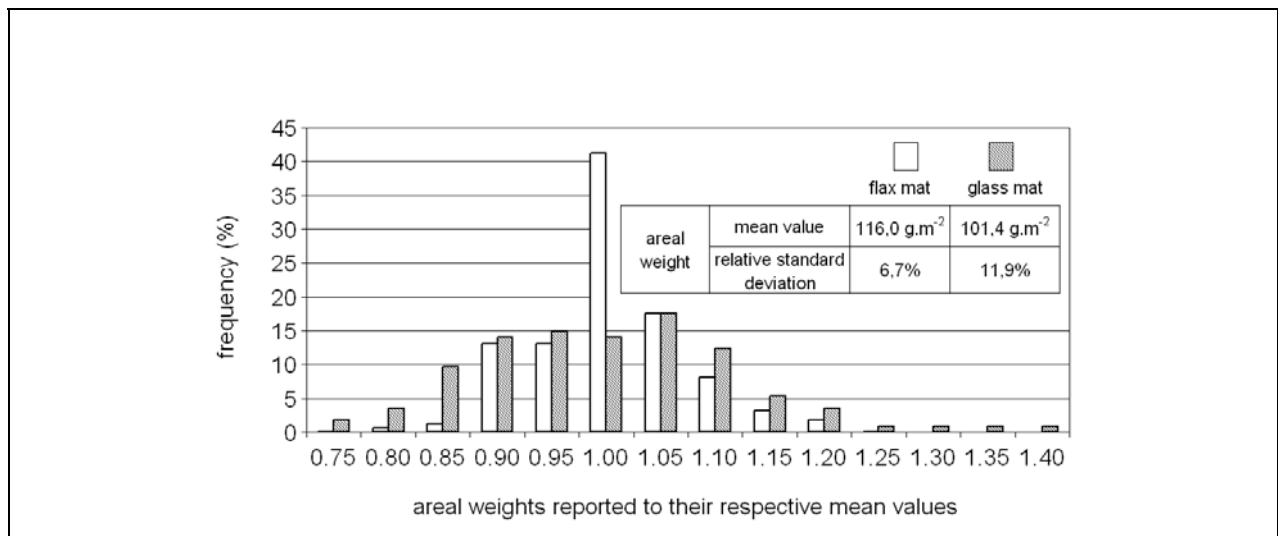

Fig. 2 Areal weights comparison between flax mats and glass mats (normalized to their respective mean values).

TRANSVERSE PERMEABILITY MEASUREMENTS IN FLAX MATS PREFORMS

Experimental Procedure

The permeability of a fibrous medium expresses its capacity to let a fluid flow through the free space between the fibers. This free space is called porosity. Permeability is homogeneous to a surface and is expressed in m^2 units. Permeability measurements are performed on a stack of twenty samples of flax mats. Flax mat samples were described in the previous section and the experimental permeability device is presented in a former publication [6]. The fluid used is a silicone oil with a viscosity around 0.1 Pa.s. Silicone oil flows at a constant velocity perpendicularly to the mats compressed to a defined thickness. As permeability measurement is achieved from a totally wet preform, one can refer to transverse saturated permeability, K_{sat}^z.

Flow pressure gradient is measured over the whole preform then K_{sat}^z values are deduced using Darcy's law [5]. The volume fraction of fibers, V_f, associated to each permeability measurement, is calculated from the preform thickness by:

$$V_f = \frac{20\,M_S}{\rho\,e} \tag{1}$$

where M_S, the areal weight of mats ($M_S = 116$ g.m^2) and ρ, the density of flax ($\rho = 1.54$ g.m^3). After the measurement of the permeability at a given fiber volume fraction, the compression over the preform is increased. As a result, the thickness of the preform is reduced and the volume fraction of fibers increases. The interesting range of volume fraction of fibers is scanned that way by increments. Here is the advantage of this permeability measurement device: transverse permeability values are obtained for the whole range of volume fractions of fibers from only one preform during one experiment.

Transverse Saturated Permeability Results

Permeability values are reported in Fig. 3 for two experiments. The two series show only small differences, so one can conclude that the measurement procedure gives reproducible results. Permeability values decrease as the volume fraction of fibers increases. Transverse saturated permeability for flax mats presents high values (more than 10^{-11} m^2 as far as the volume fraction of fibers is about 28%). The high porosity of the flax mats as shown in Fig. 1 is directly linked to these high permeability values.

TRANSVERSE SATURATED PERMEABILITY INTERPRETED BY KOZENY-CARMAN RELATION

Kozeny-Carman relation

Various studies deal with the hydraulic resistance of disordered fibrous media. The saturated permeability of fibrous media is often interpreted by the Kozeny-Carman relation. This relation can be written [5]:

$$K_{sat}^{z} = \frac{1}{k_K \, S_0^{\,2}} \frac{(1-V_f)^3}{V_f^{\,2}}$$

(2)

where k_K is the Kozeny constant and S_0 is the specific surface area between the fibers and the fluid per unit volume of fibers and expressed in m^2/m^3 of fibers. Both parameters are to be determined.
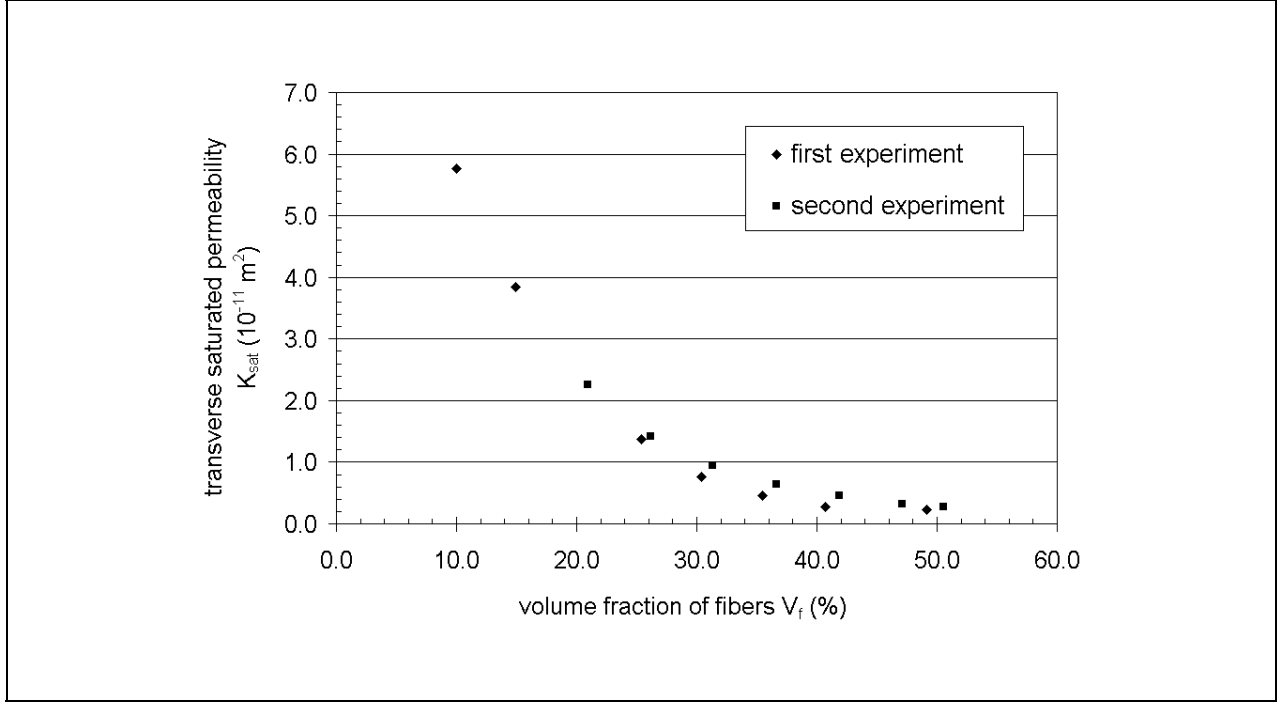

Fig. 3 Experimental values of transverse saturated permeability as a function of volume fraction of fibers for flax mats performs.

Determination of the Specific Interface Area S_0

Segments of fibers have been extracted from the flax mats, oriented parallel to one another and gathered together in a polyester matrix in order to measure the specific interface area S_0. From a plane section through the sample and an image treatment, 1227 values of perimeters and areas of fiber sections were obtained; their distributions are given in Fig. 4b. Examples of the fiber sections used for measurement are presented in Fig. 4a. The specific interface surface S_0 is deduced from the data by:

$$S_0 = \frac{\bar{s}_f}{\bar{v}_f} = \frac{\bar{l}_f}{\bar{a}_f} = \frac{95,6 \, \mu m}{495,6 \, \mu m^2} = 192900 \; m^2/m^3$$

(3)

where $\bar{s}_f$ and $\bar{v}_f$ are respectively the average area and the average volume of a fiber, $\bar{l}_f$ and $\bar{a}_f$ are respectively the average perimeter and the average area of a transversal section of a fiber.

Determination of Kozeny Constant

As K_{sat}^z values as a function of V_f (Fig. 3) and S_0 are both known, the Kozeny constant can be easily calculated from Eqn. 2. Values of Kozeny constant as a function of V_f are reported and compared in Fig. 5 to values from the literature for randomly scattered fibers [5; 7-9]. Details are given in Tab. 1 about the experimental conditions corresponding to the literature data.

Kozeny constant k_K for flax mat preforms decreases when V_f increases as previously observed in the literature. But our experimental values are higher than most of the literature data with a shift towards the highest values of V_f. The "plausible" value of 5.0 for Kozeny constant according to Carman [4] is only reached when $V_f \geq 47\%$. However, our experimental values of Kozeny constant agree fairly well with Rose's ones. These latter values are excerpt from Carman's book [5] and refer to wool media for V_f between 10% and 50%. No more detail is available on this.
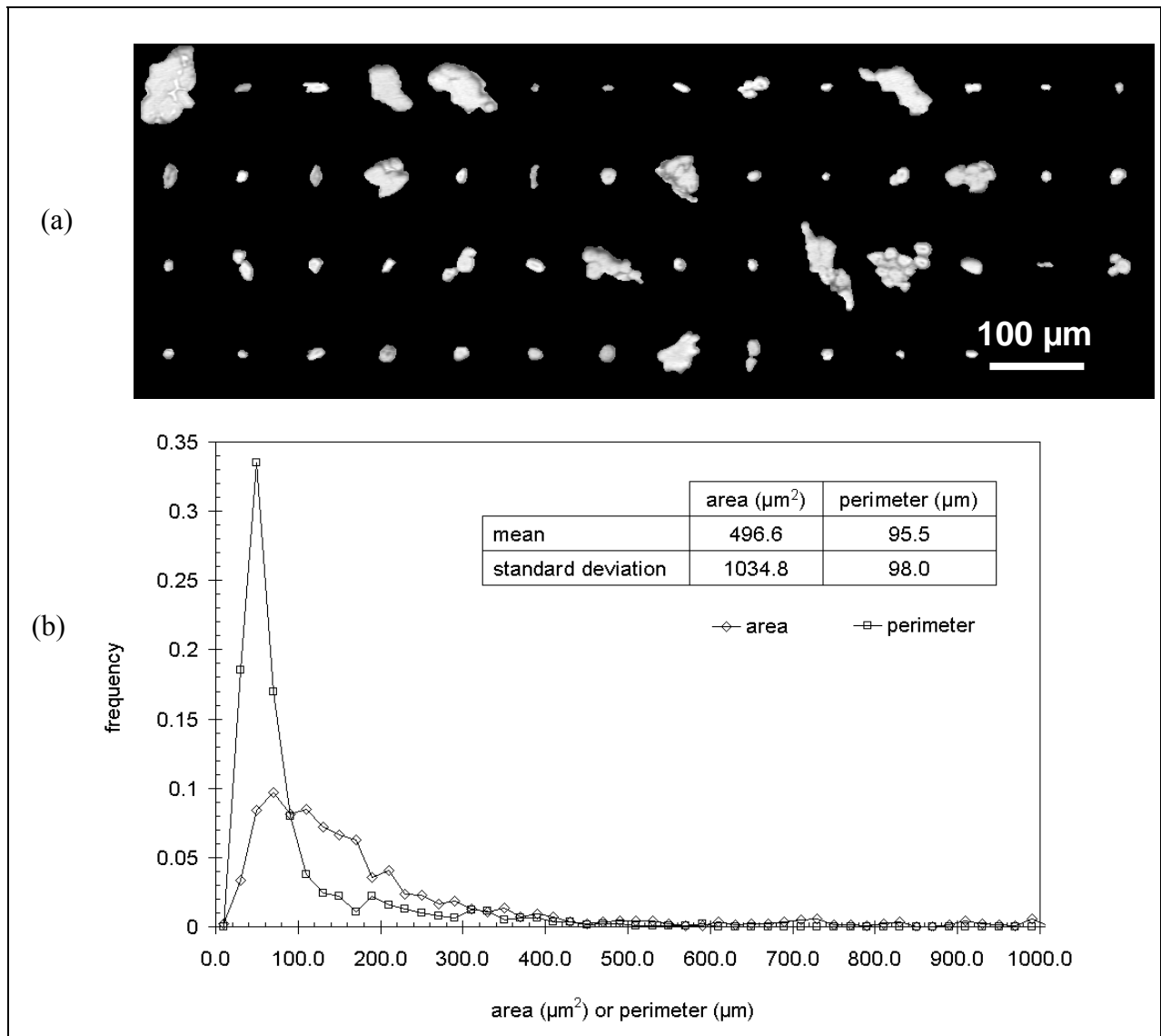

Fig. 4 Examples of flax fibers sections (a); and distribution of areas and perimeters of all fibers sections (b).

Beyond uncertainties on permeability experiments (differences between values of Kozeny constant arising from our first or second experiment are visible in Fig. 3 and 5), the major reason for the discrepancy between our values and the literature could be explained by the heterogeneity of the technical fibers (cf. Fig. 4). This heterogeneity is directly transferred to the pore network. For example, the selection of smallest fibers only for which $\bar{a}_f \leq 165$ µm^2 and $\bar{l}_f \leq 235$ µm leads to a specific interface area of 746000 m^{-1}, which is almost a fourfold increase. Measuring permeability of flax mats possessing higher specific interface area would be highly interesting in order to observe if Kozeny constant would then become closer to published values. This could confirm the hypothesis of the importance of heterogeneity for technical fibers.

Following Eqn. 2, at a given volume fraction of fibers, high values of Kozeny constant lead to flax mats permeability values lower than for other random fibrous media. A modification in bundles separation during the mechanical extraction of fibers or an improvement in the mats elaboration could change the specific interface area of flax mats and the Kozeny constant.

CONCLUSIONS

It can be concluded from this work that the transverse permeability of flax mats is sufficiently high to use a "Film Stacking" process. Interpretation of transverse permeability values by Kozeny-Carman relation shows that the Kozeny constant is higher for the studied flax mats than for other random fibrous media. The large size distribution of the technical fibers seems to be at the origin of this phenomenon. The transverse permeability could be increased by a modification of the hackling process.
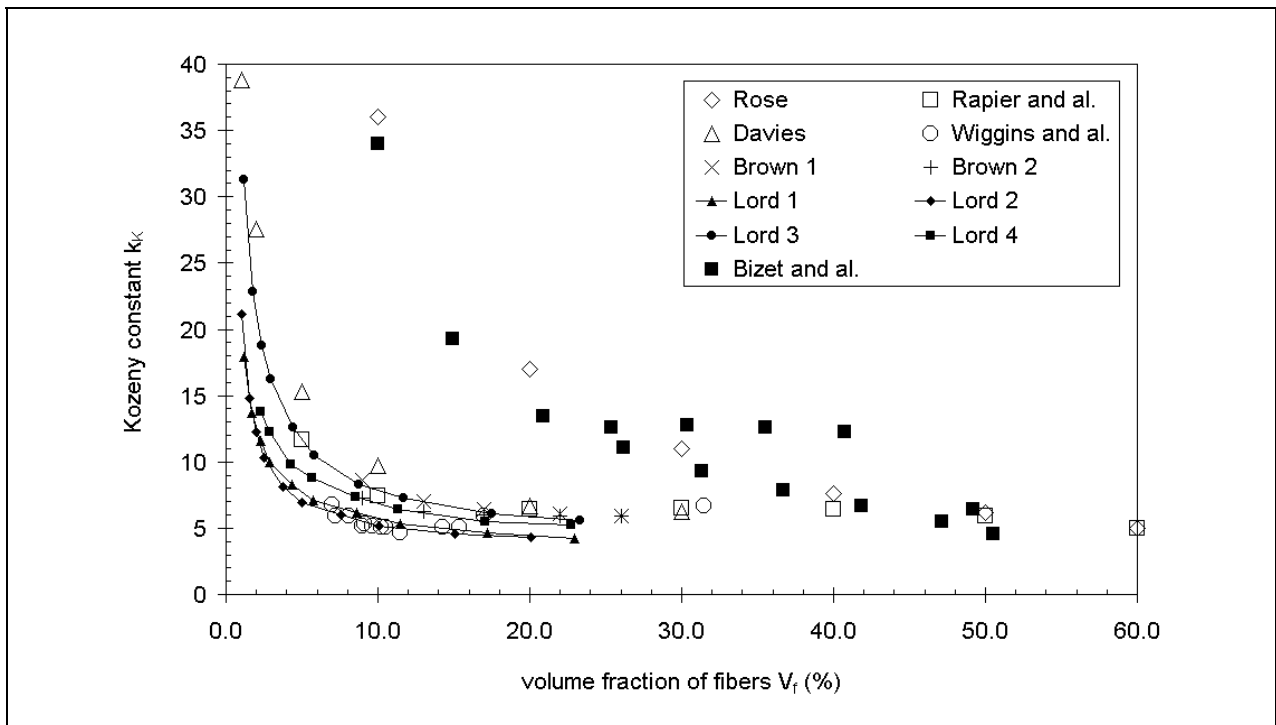

Fig. 4 Kozeny constant values compared with literature data.

Table 1 Experimental details for some Kozeny constants found in the literature

Reference			nature of fibers	nature of fluid	S_0 (m^{-1})	remarks about spatial arrangement
Rose	[5]		mostly lead wool	?	?	?
Rapier and Duffield	[5]		?	?	?	?
Davies	[5]		beds of fibres used for air filters	?	?	more or less random but fibers probably tend to lie normal to the direction of flow
Wiggins, Campbell and Maas	[7]	1	glass fibres in 400 µm diameter and 0.8 mm length	water	11500	fibrous materials cut into lengths / random-packed beds (without other precision)
		2	copper wires in 101 µm diameter and 5 mm length	water	39400	
		3	glass wool in 1.5 and 6 mm lengths	water benzene	220000	
		4	fibre glass in 1.5, 8, 10 mm lengths or merely packed	water benzene	700000	
		5	50 % glass wool and 50% fibre glass in 1.5 mm lengths	water	471000	
		6	celanese yarn of 5 mm length	benzene	~300000	
Brown	[8]	1	wool filtering glass Pyrex fibers	air	436050	thin sheets are cut in discs, laid on top of the other so fibers are oriented essentially perpendicular to the tube axis
		2				same fibres cut into 1 cm length and placed with no preferred orientation but truly randomly orientation is "doubftul"
Lord	[9]	1	cotton	air	~360000	plugs of fibers packed in a random arrangement but compression tends to orientate fibers predominantly in horizontal planes across the holder
		2	viscose rayon		~255000	
		3	wool		186000	
		4	silk		411000	

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