1 Introduction

Ultrasound is mechanical pressure waves (or sound waves) with a frequency greater than the upper limit of human hearing, approximately 20 kilohertz, what is commonly known. Sound waves of very high frequency – above the limit of human hearing – are known since the end of XIX century. They were first generated by Galton whistle (Francis Galton, England 1876), and first techniques of echo-sound based on piezoelectric effect (brothers Curie, France 1880) were designed.

Nowadays this physical phenomenon is mainly applied in engineering and medical diagnostics. In textile and clothing industries ultrasound is used for bonding of different types of materials, for welding of free ends of materials, for quality inspection of composites mainly. New trends of ultrasound implied in textile technology processes are looking for. The research applies to the study of ultrasound wave motion over a liquid, and a change of liquid’s properties under the influence of formation of ultrasound cavitations. So, new techniques that are developing nowadays are: (i) the application of ultrasound in textile pre-treatment and finishing processes [1]; (ii) using the ultrasound treatment for decreasing of a viscosity and a surface tension of resin systems, which take place in a composites production [2]; (iii) impregnation of nonwoven webs with binders and polymer solutions [3]; (iv) using of ultrasound in nanocomposite materials production for nanofiber webs impregnation with different matrixes, for uniform distribution of resin and for removal of air bubbles presented inside the nanofiber webs [3-5].

The theoretical description of ultrasound wave motion is based on acoustics’ laws and contains a composition and resolving a set of difficult differential equations. From the end of 1980’s researchers stated to work on a problem of computer simulation of sound wave propagation after a relatively simple simulation model was designed. It was the FHP lattice gas model based on a cellular automata method (CA) intended for dynamical process’s studies. From literature overview [6] was detected, that researches attended to simulation of circular and plane waves mainly, the attenuation of plane waves for different frequencies and low density of lattice gas was studied. It was not found any kind of information about computer simulation of high frequency wave motion through the porous media.

So, presenting article doesn’t contain experiments with manufacturing of composite materials reinforced by electrospun nanofibers only but a new interesting possibility how to simulate the action of high frequency waves is presented.

2 Computer simulation of high frequency waves

Creation of simulation models and computer simulation of physical systems and physical processes is a modern efficient method for study and improvement of their behavior. Usage of simulation methodology gives a whole range of advantages. Computer simulation enables to estimate, examine and consider behavior of physical system in different situations and under various conditions. It is known that mathematical models are often too complicated and hard to solve or on the contrary too simple and inadequate to reality. So, in many cases it is better to monitoring the behavior of real dynamic physical system by means of computer simulation instead of complicated mathematical modeling or costly experimental observations [7].
We are interesting in transmit of ultrasound in liquids. Being a sound wave, ultrasound has the same physical substance, where the movement of the vibrating body is communicated to the molecules of medium. In this case electrical energy is transmitting into mechanical and then into acoustic. Many types of waves exist in a real world. But the biggest part of waves counts to be harmonic plane waves, that is simplify for mathematical description. So, an own simulation model for study high frequency harmonic waves generation and propagation in a free cavity with fluids of different densities and through the porous media with different porosity was designed as a first step. Our idea was to base the high frequency wave’s simulation on a well known equation of harmonic propagation. The main part of simulation program is described in [8].

2.1 Basic information about simulation method and model which were used

Our simulation program is based on a two-dimensional FHP-II lattice gas model. The FHP-II model was developed in the end of 1980’s on a basis of cellular automata method and a simple FHP model; it is falling to the broad set of lattice gas models. Cellular automata method offers the main definitions like: cell, cell’s state, finite automata, neighborhood, etc., which are using in FHP lattice gas models too. The simple FHP model consists of an ensemble of fluid “particles” which move on an underlying triangular Bravais lattice. Each particle moves along one of the six link directions, which are given by lattice geometry (see Figure 1). Particles are moving at regular time intervals (time steps) from node to node, and can be scattered by local collisions according to a node-independent collision rules. Only one particle can travel along one link. FHP-II lattice gas model has the same properties as the simple FHP model but includes the possibility of one rest particle per node in addition to the six moving particles of the simple FHP model.

2.2 Computer simulation results

Our first simulation experiment was focused on a detection of wave attenuation in the free cavity in respect of different frequencies and densities of liquid media (See Figure 2).

We tried to detect if our simulation model is in a good agreement with a theoretical knowledge. It is known that attenuation is proportional to the total viscosity of the liquid and that is increases with the square of the frequency.

In the FHP model viscosity is done by a density of particles per unit volume (per site or per node) and type of collisions. Densities, which were used, are 3.5 and 1.2 particles per node. Time – is a discrete variable in the simulation model and during one time step (t.s.) have to take place an one change of a cell’s state only. Therefore it was used a period (which is inverse proportional to the frequency) instead of frequency. How to calculate the attenuation from simulation outputs is described in [9]. Figure 3 shows computer simulation results, where the dependence between attenuation of waves, period and density is presented.

Our simulation results are in a good agreement with the theoretical knowledge. It is possible to see that
attenuation decreases with the square of the period. The similar experiment was realized in [10] and similar results were obtained.

We are planning to apply our simulation model for study of wave motion through different porous media. Figure 4 shows first computer simulation result of wave motion through the regular porous media with different porosity presented by a grey rectangle at the picture. It is evident that absorption of wave depends on a structure of porous media. Structures, where porous size is close to a mean free path of molecule, are good absorbers of sound waves. These structures can be present by nanoporous materials in a real scale. And for us it would be interesting to compare this result with a real experiment in future.

3 Composites with electrospun nanofibers by means of ultrasound

3.1 Literature review

Nanofibers are generally fibers of diameter smaller than 1 μm. Several methods that are used to prepare nanofibers were described during last few years. Those methods are based on different principles: (i) fibers formed in a spinneret are drawn by a stream of hot air (also known as melt-blown method); (ii) dissolving polymer matrix of so called islands-in-the-sea bicomponent fibers; (iii) electrospinning. This article is focused on nanofibers produced during electrospinning process. Electrospinning method has been described in several patents [11-13]. It seems to be the most important method among the processes leading to nanofibers, although some companies obviously develop production methods based on different principles [14]. Another method with higher efficiency of nanofiber production was developed recently at the Technical University of Liberec at the Department of Nonwovens [15] and patented [16].

There are generally known advantages of using traditional polymer fibers as reinforcement material for production of composites. Needles to say, nanofibers will also eventually find important applications in composite materials making. Nanofibers can have even better mechanical properties than micro fibers of the same raw; this is because fibers are being elongated in electrostatic field and as a result there are less of defects per unit length of nanofibers [17]. The enlargement of surface area means greater possibility to stop the progress of defect. The enlargement of fibers surface also leads to creation of more functional groups at the surface and consequently to increase of amount of transitions between matrix and reinforcement.

Majority of articles (in the available literature) devoted to composite materials reinforced by nanofibers is focused on carbon nanofiber or nanotubes (see examples [18-21]). It is necessary to note here that possibilities of carbon nanofibers producing from polymer nanofibers precursor produced by electrospinning exists [22-25]. Such research aimed at carbonization of PVA electrospun nanofiber layer is being developed at our department too. Only sporadic papers are dealing with manufacturing of composite materials reinforced by polymer nanofibers produced by electrospinning. In 1999 Kim and Reneker [26, 27] studied reinforcing effect of polybenzididazol (PBI) nanofibers produced by electrospinning in epoxy resin and rubber matrix. Bergshoef and Vancso [28] produced composite material reinforced by nanofibers PAD-4.6 obtained from electrospinning with epoxy resin as a matrix in 1999 also. The appreciable improvement in mechanical properties of reinforced material compared to solitary matrix was registered in both of events in spite of very low weight fraction of fibers in composite (3-15 %). In latter research the transparency of composites reinforced by nanofibers was employed.

Concerning the improvement of stiffness and strength, researchers also tried to modify other mechanical properties of composites by using electrospun nanofibers. The particularity of nanofiber material is its very high surface area to volume ratio. This property of nanofiber materials may be advantageous with the aim to increase the resistance to delamination of composite laminates. This information came from US patent [29]. Authors of the patent (Dzenis & Reneker) used polymer electrospun nanofiber layers between laminates (the ones consisting of classical fibers used as a composite reinforcement material) of a composite. The results, which were obtained, showed the improvement in delamination resistance. In other terms
we can say that the interlaminar toughness of such type of hybrid composites was improved. Electrospun nanofibers reinforced polymer composites have been developed mainly for providing some outstanding physical (e.g. optical and/or electrical) and chemical properties while sustaining their appropriate mechanical performance.

There is also necessary to have some knowledge of nanofibers orientation in electrospun nanofiber web; and also about the guiding, regimentation and orientation of these fibers to be able to design and predict the properties of result composites. From the literature review it was found that several research teams have studied possibilities of electrospun nanofiber alignment but none of them did the complete examination or used their methods for bigger amount of nanofibers. Five following techniques listed below are some of overall possible techniques which have been attempted to align electrospun nanofibers [20]: a cylinder collector with high rotating speed, an auxiliary electrode/electrical field, a thin wheel with sharp edge collector, a frame collector, a multiple electrostatic field technique.

3.2 Producing of composite materials

Nanofiber nonwoven layers made at our department up to now fulfill basic assumptions for their employing in composites. The length of fibres is far longer than critical length of fibers in composites and so system acts as endless fiber system, fibers are straight and they can be partially oriented with using additional devices, fibers are easy chemical modified (See Fig. 5). It is well known, that the yield strength of materials increases as the characteristic length of material elements (grains or fiber diameter) is made smaller, to provide more batters for dislocation or crash movement. That is why we place our hopes on the development of nanofiber-reinforced composite materials, which moreover previously mentioned strengthening effect could provide us with extremely thin and hence flexible plates.

First attempts with nanofiber-reinforced composites were based on the system composed of polyvinyl alcohol fibers and epoxy matrix. The Hand Lay-up fabrication method was used. There were two limits in this procedure: the content of reinforcing nanofibers in such composite was extremely low – estimation is about 2.3% by volume, air trapped in the resin was not possible to get out from composite. This is why we decided to focus our research to improvement of our fabrication towards magnification of content of nanofibers in composites without bubbles. So, we decided to use ultrasound for better impregnation of nanofibers and for removing of air bubbles trapped inside wetted material. Our decision was based on the knowledge, that the application of ultrasound enhances the sorption of highly viscous liquid matrices [19]. Especially the using of ultrasonic treatments significantly decreased the viscosity and surface tension of the resin systems and increased the wettability of the fiber surface due to the ultrasonic cavitation effects [30]. Therefore, the using of ultrasound in the impregnation process brings the improvement of adsorption and the better escape of air from inside to outside of impregnated material [31]. For our experiments we used the ultrasonic treatment with frequency 30 kHz and power 30W. The sets up of our experiments for impregnations of nonwoven from nanofibers are shown at a Figure 6. There were used different liquids as a matrix: epoxy resin with extremely high viscosity ($60\text{Pas}$), epoxy resin with lower viscosity, polyester resin and water solution of acrylic binder in 50% concentration (the lowest viscosity one). The fiber material used in these experiments was produced at our department and had these parameters: 1) basic polymer is PVA (polyvinyl alcohol), electrospinning proceeds with addition of

![Figure 5: Picture of PVA nanofibers produced at the Department of Nonwovens (TUL) from SEM.](image)

![Figure 6: Set up of experiments for impregnation of layers from nanofibers: a) for high viscous, b) for low viscous liquids: 1 – nanofiber material, 2 – drop, 3 – ultrasonic transducer, 4 – ultrasonic generator.](image)
polyacryl acid for later cross-linking at high temperature, random orientation of fibers, surface density 3 gm², average of fibers 236 nm with standard deviation 79 nm, 2) basic polymer is PA 6.12 (polyamide), electrospinning proceeds with addition of mixture of formic acid and acetic acid, surface density 3 gm², average of fibers 279 nm with standard deviation 117 nm.

3.3 Results and Discussion

The result samples of composite materials compound from several nanofiber nonwoven layers and the epoxy resin or acrylic binder were completely transparent. The system with acrylic matrix showed a plastic foil like behaviour. The fiber contents about 15% by weight and about 5% by volume were determined for composite material nanofiber layers – epoxy resin and fiber contents about 34% by weight and 31% by volume were determined for composites nanofiber layers – acrylic binder. Basic assumptions about tensile strength tests, those composites with epoxy matrix have higher strength then composites with acrylic matrix and conversely the composites with acrylic matrix have higher elongation then composites with epoxy resin, were corroborate. Other structural, mechanical and physical properties will be presented and discussed at conference.

All these tested materials are only models. If we have optimal composition of the composites, we will start with experiments according to the patent [29], where our composites in a “prepreg” form will be used for an increasing of delaminating resistance of laminates..

4 Conclusions

Ultrasound is a very important and interesting phenomenon not only in the textile industry. Our article shows we are able to simulate the basic principles and effect of it. Also, we are able to find relatively new applications for it. Usage of ultrasound for the impregnation of nanofiber materials is “elegant” method how to accomplish higher content of fibers and better uniformity of liquid matrix allocation in the nanofiber web.

Over all these successes, the long way full of confirming experiments is still in front of us in both simulation and production of composite materials.

Acknowledgement

This work was supported by a grant of the European Commission, NMP2-CT-2003-505892-1, Ultratec.

References


[22] http://webpages.sdsmt.edu/~hfong/1.1.html


