

SMART COMPOSITES WITH INTEGRATED TINY PIEZOELECTRIC FIBERS

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SUMMARY: A new type of multifunctional composite material has been established by integrating parallel arrays of tiny piezoelectric fibers with interdigital electrodes into polymer matrices. This follows the approach of structural conformity to maintain the mechanical properties of the composite materials. The access to highly integrated materials consisting of at least four solid phases is based on the combination of fiber technology, electroding, composite fabrication and adaptive control. The sensing as well as the actuating properties of the resulting materials are evident. This offers a wide range of possible applications like active and passive vibration damping, health monitoring, structural control and impact detection.

KEYWORDS: piezoelectric fibers, sensor, impact, adaptive structures.

INTRODUCTION

Over the past 20 years, material science has been searching for novel, useful, highly integrated and efficiently working composite materials. In several approaches nature has become a model for engineers to develop a new type of materials, namely multifunctional composite materials (also called adaptive or smart materials). Effective biological structures have developed during the evolution of life. They are able to adapt efficiently to changing environmental conditions in their habitat. This ability is based on incorporated sensing as well as actuating functions, both connected by signal paths and controlled by e.g. a brain as processing unit. As soon as the sensors detect an impact, they generate a signal which is transmitted to the brain. This receiver in return sends a control signal which releases an adequate actuator reaction.

To copy biological structures is not possible at present because of the very high degree of integration, excellent efficiency of the sensor/actuator response and the capability of damage repair. Nevertheless, the idea to mimic biological principles for multifunctional materials, e.g. by combining active solid materials with polymers in well organized architectures, came up in the early 80ies [1]. As active materials, electroceramics are the most promising candidates [2].

The first approach was to tailor the properties by combining active and passive materials in defined architectures. Then, it was suggested to integrate active materials into composite materials [3]. Such „smart materials“ consist of 3 or more solid phases and can be potentially used in technical applications. They should contain sensors, actuators and controllers to make the material adaptive.

Amongst all known functional materials with useful sensor/actuator coupling the piezoceramics in the system $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT) including substitutions and dopants occupy a special position. These materials offer both sensing as well as actuating behaviour although they exhibit a relatively small strain in the range from 1 to 1.5 ‰ and need high electric driving fields of 20 kV/cm. However, they are suitable for dynamic and precise positioning, their energy density is exceeded only by shape memory alloys and biological muscles [4] and they can develop stresses up to 3 kN/cm².

The development of tiny functional components like fibers opens new opportunities in material design in the sense of structural conformity. Structural conformity means that the properties of fiber-reinforced composite materials (high specific strength and stiffness as well as high strain to failure) should not be deteriorated by the integrated functional components and their wiring. The consequence is to minimize the geometric dimensions of the functional components as long as they can maintain their functionality as well as their controllability. According to this concept the integration of functional components in form of thin fibers seems to be a promising approach.

The development of highly integrated composite materials is a matter of fusion and extension of one new and at least 4 existing technologies: the PZT-fiber preparation is a new technology, whereas electroding, composite fabrication and adaptive control have to be adjusted to new requirements.

Using this approach PZT fibers can be applied as integrated compression as well as tension sensors. Active and passive vibration damping, health monitoring and control of geometry are even more important fields of application [5,6]. Last but not least, PZT fibers with small diameters will give access to new types of ultrasonic transducers [7].

OUR APPROACH: STRUCTURAL CONFORMITY

The developmental steps from passive structures towards highly integrated adaptive structures are the following: as soon as a passive structure is affected by disturbing forces vibrations and noise will be generated. If those structures are equipped with sensors and actuators, both connected with a controller, vibrations and noise can be actively damped. However, the structure becomes heavier due to the additional external components because piezoelectric structural actuator systems are traditionally based on monolithic planar materials in stacked architectures. Structures of this type are already available on the market [8]. The next consequent step is the integration of sensors and actuators within the structure, the adaptive controller still being external. Such systems can be designed to be free of vibrations, quiet and stable, but still being light weight! In the highest degree of integration the controller is also contained within the composite. Structures of the last both types generate new requirements on the design of sensing as well as actuating components: if the mechanical properties of the

composite material are to be affected as little as possible, then their geometric dimensions have to be minimized resulting in “structural conformity”. What does this mean?

The mechanical properties of fiber-reinforced composite materials are determined by the type and content of reinforcing as well as functional fibers. In order to calculate qualitatively at which diameters the functional fibers do not interfere with the properties of fiber-reinforced composites, it makes sense to compare the elastic behaviour of fibers of different types and diameters. One useful measure are their bending moments. The bending moment M can be calculated qualitatively for cylindrical shapes - like fibers - by the formula:

$$M \approx E \cdot d^3 / 32 \quad [9]$$

where E = Young's modulus, d = fiber diameter.

The formula tells that smaller fiber diameters result in smaller bending moments. To fulfill structural conformity, it can be concluded that the bending moments of reinforcing as well as piezoelectric fibers with different diameters and Young's moduli should ideally be quite similar. As a realistic case a glass fiber-reinforced composite (GFC) can be considered: the Young's moduli of glass and PZT are comparable, consequently their diameters should not differ. Another consequence can be drawn from this consideration: in the case of carbon fiber-reinforced composites with a distinctly higher Young's modulus and a diameter of 6 μm of C-fibers PZT fibers with a diameter smaller than 20 μm should be used.

From this qualitative consideration the conclusion can be drawn that a technology is needed that allows to produce PZT fibers with a final diameter of at most 30 μm or smaller. By using such fibers structural conformity can be achieved with consequences for the improved lifespan and the reliability of adaptive composites.

The preparation and integration of PZT fibers with such small diameters require a technology which permits to handle the fibers carefully during all production steps, to align them precisely in an ordered architecture and to electrodeposit them. The electrodepositing process as such requires microtechniques which are still under development.

PREPARATION OF PIEZOELECTRIC CERAMIC FIBERS

The preparation of piezoelectric fibers with diameters $< 30 \mu\text{m}$ has been started in the 90ies [10], but is still a challenge for material technology. In literature at least three different routes have been reported. They differ essentially in the precursors used for their preparation, the achievable diameters in the sintered state and in the sintering temperatures themselves.

Fiber preparation routes based on the extrusion of polymer-supported PZT powder suspensions cannot produce final fiber diameters smaller than 125 μm [11], but they are used to develop Active Fiber Composites (AFC) for an integratable actuator [12]. Another fiber preparation route has been reported which starts with a PZT powder suspension based on a cellulose xanthogenate solution (VSSP method = viscous solution spinning process). Final fiber diameters of 20 μm have been achieved, but the necessary sintering temperature is so high ($> 1250 \text{ }^\circ\text{C}$) that the fibers sinter together and cannot be handled as single fibers [13]. The result is a highly porous aggregate of PZT “fibers” which can be used only for the development of acoustically adapted materials, e.g. for hydrophones [14]. It was recently reported that

single fiber diameters in the range of 10 to 25 μm have been achieved by the VSSP method [15].

The third route based on the sol-gel process is favourable, because in this way diameters $< 30 \mu\text{m}$ fibers sinterable at lower temperatures have been achieved [15-17]. Due to the comparably low sintering temperatures ($< 1000 \text{ }^\circ\text{C}$) it is possible to fabricate fibers which can be handled in the sintered state as single filaments.

Sol-gel derived PZT fibers have been used for the following steps [16]. The fiber stoichiometry presently used is adjusted to lead to the final composition $\text{Pb}(\text{Zr}_{0.53}\text{Ti}_{0.47})\text{O}_3$. This composition is not yet the optimum for the intended applications. A “hard” PZT material exhibiting a high coercitivity and low piezoelectric charge constant results. This means that its poling needs high electric fields up to 50 kV/cm at elevated temperatures, and a relatively small charge will be generated under compressive or tensile load.

Sol-gel derived PZT fibers show an extraordinarily high sintering activity. They can be completely densified at temperatures $< 950 \text{ }^\circ\text{C}$ (Fig. 1) and can be handled as single fibers after sintering.

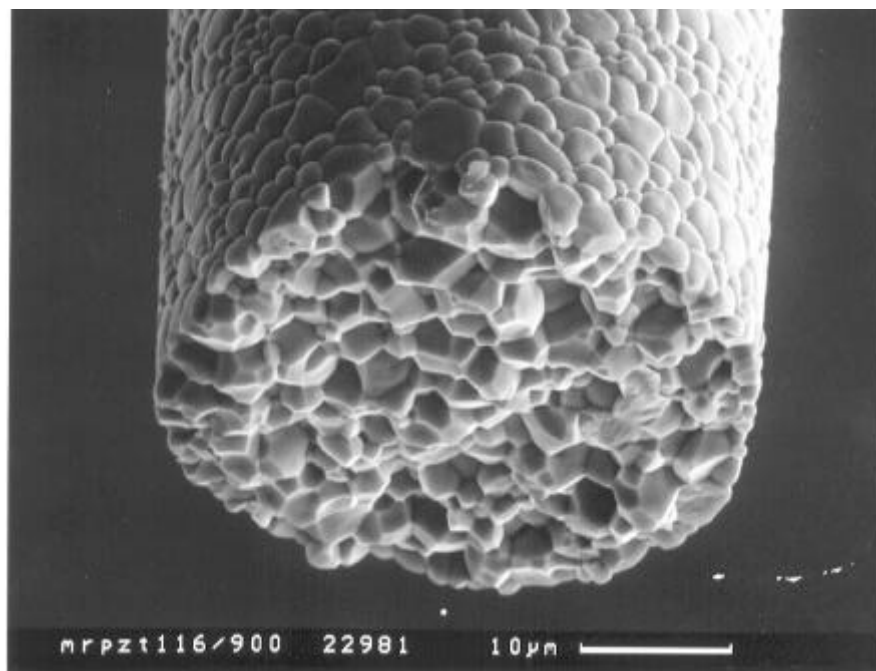


Fig. 1: SEM micrograph of PZT fibers with the composition $\text{Pb}(\text{Zr}_{0.53}\text{Ti}_{0.47})\text{O}_3$ sintered at $925 \text{ }^\circ\text{C}$ [16]

The sintered fibers exhibit the targeted stoichiometry, the microstructure is pore free and consists of grains between 2 and 4 μm in diameter. The measured tensile strength is in the range of up to 300 MPa, Young's modulus was estimated to be 70 GPa.

PZT fibers are not suitable to take over load bearing functions within the composite material. Their elastic properties cannot be improved for structural reasons, the tensile strength is - among other facts - a function of the failure probability within the microstructure, particularly

on the fiber surface. For electromechanical reasons a coarse microstructure is preferred leading to a grooved fiber surface. These grooves are sources of local stress concentrations. Due to these reasons the fiber strength is limited.

Conclusions can be drawn concerning the structural arrangement of composite materials with integrated and electroded PZT fibers: the resulting material will consist of at least four phases. The single components are the polymeric matrix, the reinforcing fiber (glass or carbon), PZT fibers and their electrodes.

ELECTRODIZING AND INTEGRATION

The process route to active fiber laminates is shown in Fig. 2. The structure of the electrodes has to allow that unidirectionally (UD) aligned PZT fibers can be poled and then addressed. The fiber integration should affect the structure of the composite as little as possible. The access to the internal electrodes is necessary to allow the in- and output of charges.

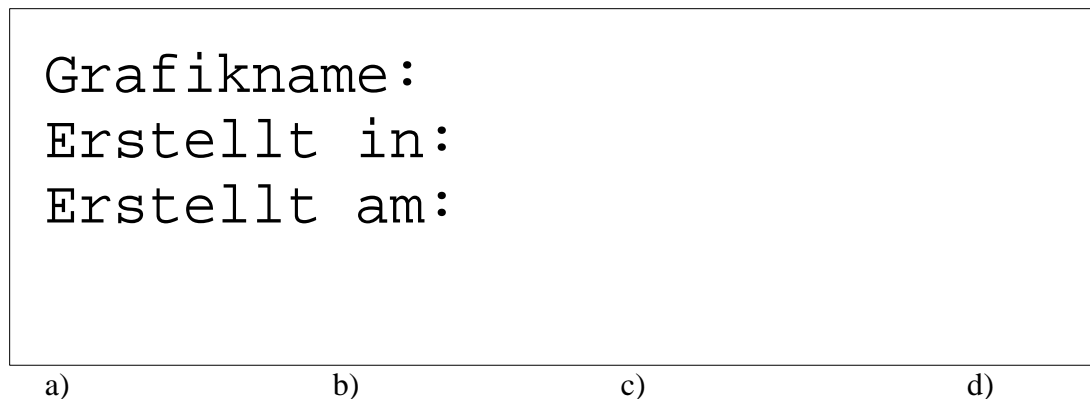
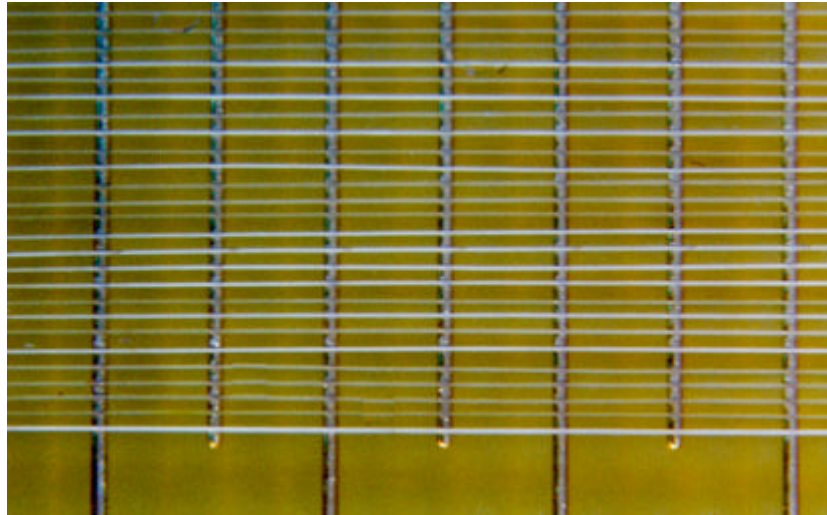


Fig. 2: Scheme of a fiber reinforced, laminated composite with integrated PZT fiber single sheets

*a) electroding of the UD piezofiber single sheet, b) electroded piezofiber single sheet
c) piezofiber single sheets between GFC or CFC sheets, d) active laminate with electrodes*

The use of active single sheets can ensure a high degree of structural conformity but generates high demands on the PZT fiber technology. The gel fibers have to be precisely aligned in UD architectures; this geometry is not allowed to change over the whole process. It has to be taken into account that a linear shrinkage of the fibers of about 40 % occurs due to pyrolysis/oxidation and sintering. Sintered PZT fibers in aligned UD geometry can now be produced by careful process control. Only very small deviations within the fiber array can be tolerated, because the width and thickness of the electrodes as well as the distance between the electrodes must be kept with a tolerance below 5 %. In Fig. 3 a real fiber/electrode assembly is shown.



*Fig. 3: Real PZT fiber/electrode structure, electrode distance = 1 mm
(preparation: Fraunhofer IFAM)*

The piezoelectric fibers in this array successfully have been poled. The sheets can be integrated on or between GFC sheets.

PROPERTIES OF COMPOSITES WITH INTEGRATED PZT FIBERS

PZT fibers as integrated sensor

PZT fibers as described above were integrated within single sheets with a volume content of about 30 %. Such active sheets have been embedded in a GFC plate with a dimension of 20 x 20 cm². After that the PZT fibers can be poled with electric fields between 30 and 50 KV/cm at a temperature of 120 °C and behave anisotropically, as expected. As soon as the embedded PZT fiber array comes under a mechanical load charges are generated. The complete GFC plate is covered by a net of 81 measuring points in a regular manner. A mechanical load of 14.3 g has been dropped from a height of 15 cm on each of these points resulting in an impact energy of 0,02 Nm. The impacts generates a figure of measured charges as shown in Fig. 4. This experimental evidence demonstrates that impact sensors can be built by tiny, integrated PZT fiber/electrode arrays.

A sensitivity of about 40 pC/N was calculated from the measurement of the generated voltage. The need for the change to a “soft” PZT composition as mentioned above is now evident. The charges generated are directly proportional to the piezoelectric charge constant d_{33} (d_{33} (hard) 220 pC/ N, d_{33} (soft) 600 pC/ N). With the change to soft PZT fibers the sensitivity of the sensor will be nearly tripled.

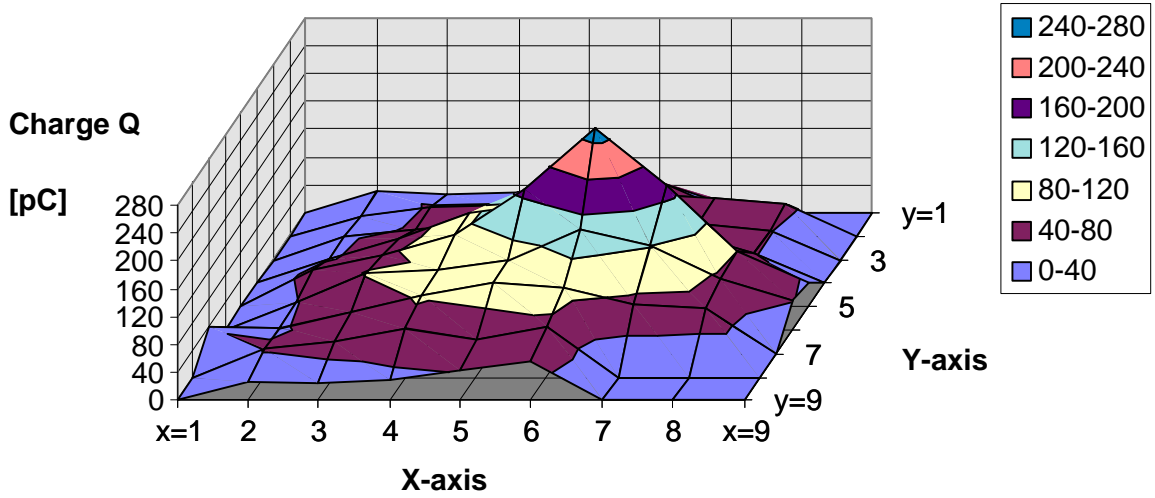


Fig. 4: Charge diagram measured on a GFK sheet (20 x 20 cm²) which contains poled PZT fibers after local impacts with an energy of 0,02 Nm (measurement: Fraunhofer IKTS)

PZT fiber architectures for passive vibration damping

The same PZT fiber architecture as shown in Fig. 2d also allows the damping of mechanical vibrations. As soon as poled PZT fibers in such arrays undergo compression or tension they generate charges by the direct piezoelectric effect. These electrical charges can be collected by the electrodes and can be transferred to Joule heat by shunting. By this mechanism mechanical energy is taken away from the system: it will be damped.

Derived from theoretical calculations of the possible energy transfer, comparably high damping values can be expected. Most probably damping of frequencies in the range of up to 2000 Hz will be reached. The maximum energy dissipation achievable, which is caused by a shunt after transformation of mechanical in electrical energy, can be calculated as follows:

$$\sin \delta = k^2 / 2 (1 - k^2)^{1/2} , \text{ with } \omega \epsilon^\sigma \rho = 1 / (1-k)^{1/2} \quad (2) \quad [19]$$

with: k = electromechanical coupling coefficient
 ω = frequency
 ϵ = permittivity at constant mechanical stress
 ρ = specific resistance

The achievable damping depends on the coupling factor of the piezoceramic ($k_{33} = 0.6$), the degree of coupling between the array and the source of vibration and the frequency used. At a frequency of 200 Hz the value varies between 8 and 22 % per single vibration (Fig. 5). The experiment is in good agreement with the calculated form of the dissipation curve (Fig. 5, cross marks).

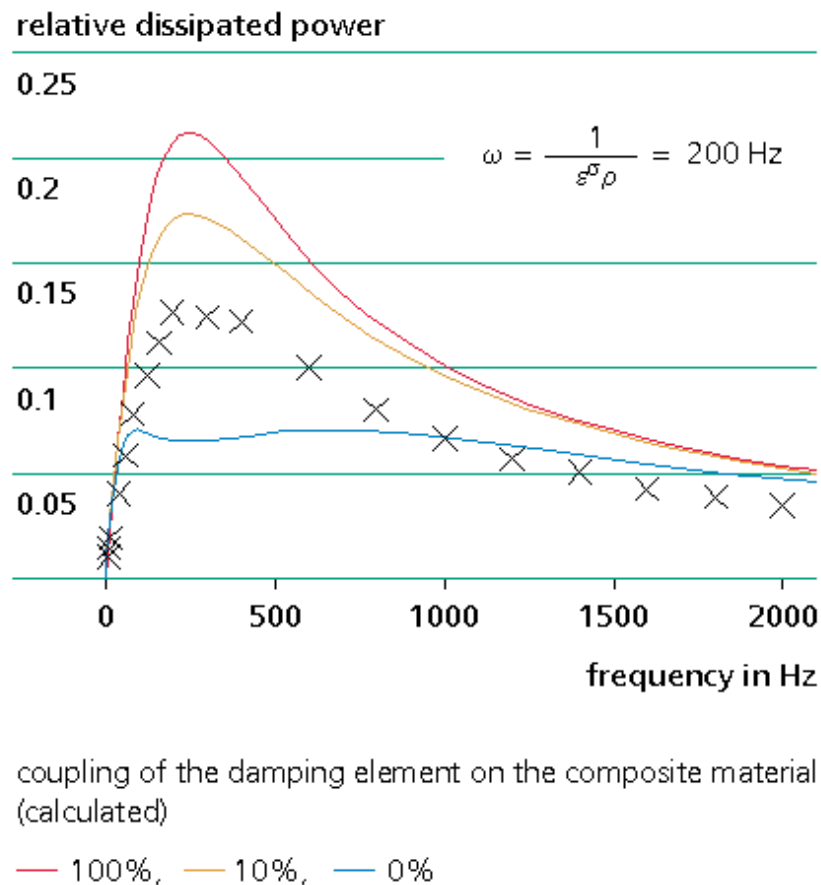


Fig. 5: Damping with integrated PZT fibers at a frequency of 200 Hz and different couplings between damping array and the vibrating exciter in comparison with a measurement (x) on a PZT fiber array (calculation: Dr. W. Kreher, TU Dresden, Germany, measurement: Fraunhofer IKTS)

The application of arrays, that can be integrated into composite materials in structural conformity, is now a matter of optimization of materials as well as the related technologies. This concerns the fibers themselves - here again the change of composition to a soft system is necessary - as well as their integration and control into devices.

Integrated PZT fibers as actuator

PZT fiber arrays as shown in Figure 3 can also be used as actuators. This has been demonstrated using a glass fiber-reinforced polymer (GFR) sheet with a laminated PZT fiber array. By applying an AC signal to the PZT fiber array the sheet is excited to vibrate audibly in its resonance frequency. The effective power P of the excited vibration mode was taken as the physical measure of electromechanical coupling. Figure 6 shows the measured function $P(f)$ of a vibrating sheet ($5 \times 12 \text{ cm}^2$) driven by the integrated piezofiber transducer at an applied voltage of 2000 V (AC). The plotted function shows that at 400 Hz an effective amount of energy is exciting the vibration of the sheet in its characteristic vibration.

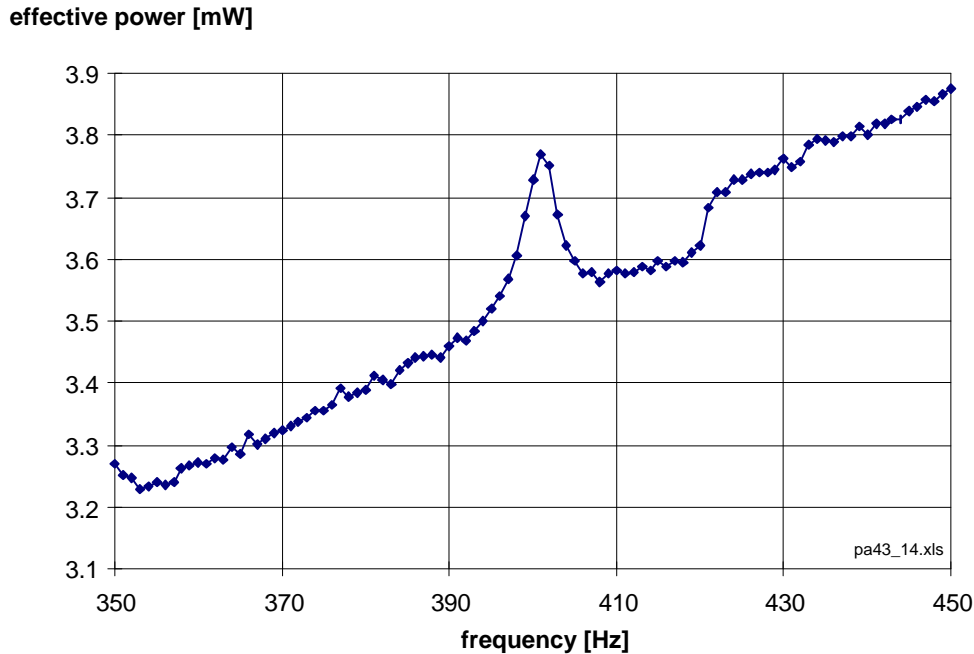


Fig. 6: Plotted effective power in dependence of the applied frequency measured in a sheet which is driven by the integrated piezofiber transducer. At 400 Hz the characteristic vibration of the sheet is shown. (measurement: Fraunhofer IKTS)

This is the first experimental evidence of an actuatoric function of PZT fiber arrays. This can be seen as the starting point of material development opening wide fields of applications like active vibration damping as well as integration of actuatoric components into composites while maintaining their structural conformity.

CONCLUSIONS

The integration and control of PZT fibers with tiny diameters into composite materials leads to a new type of material. Tiny PZT fibers can be aligned in an array, electroded with interdigital electrodes and then integrated. Evidence of sensoric and actuatoric properties of integrated PZT fiber architectures has been presented. The results achieved so far are a basis to explore the potentials of such materials by optimizing the fiber composition, the structural design of composite materials and - last but not least - the “intelligence” of the controller.

The fiber development is not yet finished. The next necessary steps are the optimization of the chemical composition and the microstructure with respect to the mentioned applications and the further development of the fiber preparation technology. Soft PZT formulations are now under investigation and will be available in the near future.

The fiber technology is still challenging. Our mid-term target is to develop and to establish a continuous process which can deliver larger quantities of fibers necessary for the development of components. At present the fibers are produced batchwise on a laboratory scale. We expect that “intelligent” composite materials with integrated PZT fibers can trigger the production of a large number of new products.

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