biblio.ugent.be

The UGent Institutional Repository is the electronic archiving and dissemination platform for all UGent research publications. Ghent University has implemented a mandate stipulating that all academic publications of UGent researchers should be deposited and archived in this repository. Except for items where current copyright restrictions apply, these papers are available in Open Access.

This item is the archived peer-reviewed author-version of:

Thin and Flexible Polymer Photonic Sensor Foils for Monitoring Composite Structures

Jeroen Missinne, Nuria Teigell Benítez, Alfredo Lamberti, Gabriele Chiesura, Geert Luyckx, Marie-Aline Mattelin, Wim Van Paepegem, Geert Van Steenberge

In: *Advanced Engineering Materials*, Volume 20, Issue 2, page 1701127 (5 pages), 2018.

<https://doi.org/10.1002/adem.201701127>

To refer to or to cite this work, please use the citation to the published version:

Jeroen Missinne, Nuria Teigell Benítez, Alfredo Lamberti, Gabriele Chiesura, Geert Luyckx, Marie-Aline Mattelin, Wim Van Paepegem, Geert Van Steenberge (2018). "Thin and Flexible Polymer Photonic Sensor Foils for Monitoring Composite Structures." *Advanced Engineering Materials* 20(2). 1701127. 10.1002/adem.201701127

DOI: 10.1002/((please add manuscript number))

Article type: Communication

Thin and flexible polymer photonic sensor foils for monitoring composite structures

Jeroen Missinne, Nuria Teigell Benítez, Alfredo Lamberti, Gabriele Chiesura, Geert Luyckx, Marie-Aline Mattelin, Wim Van Paepegem, Geert Van Steenberge*

Prof. Dr. J. Missinne, Dr. N. Teigell Benítez, M-A. Mattelin, Prof. Dr. G. Van Steenberge
Ghent University and imec, Centre of Microsystems Technology (CMST),
Technologiepark 15, Gent-Zwijnaarde, B-9052, Belgium
E-mail: jeroen.missinne@ugent.be

Dr. A. Lamberti, Dr. G. Chiesura, Dr. G. Luyckx, Prof. Dr. W. Van Paepegem
Ghent University, Department of Materials, Textiles and Chemical Engineering (MaTCh)

Keywords: Bragg gratings, composites, foil, multi-axial strain sensors, polymer waveguides.

Abstract

Thin and flexible photonic sensor foils are proposed, fabricated and tested as a promising alternative for monitoring composite structures. Sensor foils are implemented using two different optical polymers and as such optimized for multi-axial sensing and embedding within composite materials respectively. It is first shown that those sensor foils allow multi-axial strain sensing by multiplexing a multitude of Bragg grating sensors in a rosette configuration. Secondly, those sensors can be realized in very thin foils (down to 50 μm) making them suitable for embedding in composite materials during their production. This was proven by visually inspecting and by testing the functionality of the embedded sensors. Finally, owing to their low Young's modulus and flexibility, polymer sensor foils can be bent to small curvature radii and withstand large elongations. Herein, the sensors are bent down to a radius of 11 mm, and elongated by 1.4% without losing functionality.

Composites are increasingly being used in mechanical structures by virtue of their high strength- and stiffness-to-weight ratio.^[1] These materials consist of reinforcement fibers (typically glass or carbon) bonded together by a matrix of polymer resin (e.g. epoxy). Because of the presence of the reinforcement fibers, composites are inherently anisotropic, which

makes their modelling more complex. Consequently the prediction of their failure mechanisms becomes more difficult. However, this is required to avoid costly overdimensioning or premature replacement of structural parts. For this reason, feedback from sensors is vital for in situ measuring of the stress/strain state and as such monitoring the structural condition of the composite. Strain gages are traditionally being used for this purpose, but more recently optical fiber Bragg grating (FBG) sensors are increasingly being used for their immunity to electromagnetic interference, high sensitivity and multiplexing capability.^[2] Furthermore, since these sensor fibers have the same shape as the reinforcement fibers (with only a slightly larger diameter), they can be integrated inside the composite without disturbing its structural integrity, which allows measuring the actual strain at critical locations in the structure and shields them from the sometimes harsh operating conditions. The current commercially available fiber sensors, however, are mainly sensitive to strain along their longitudinal direction, and therefore, multiple precisely oriented fiber sensors are required for measuring the complete strain field.

Herein, we demonstrate the potential of a new approach using thin polymer foils in which a multitude of optical sensors in different orientations can be defined and which can be thin enough to be embedded inside composite materials. Owing to their flexibility and low Young's modulus, polymer-based optical foils can be bent down to small curvature radii (in the order of millimeters) and can withstand large elongations (up to several tens of percents for some polymers) without losing functionality.^[3]

With this approach, the mutual orientation between individual sensors in the same foil can precisely be defined during fabrication, allowing the multiple-sensor foil as a whole to be easily integrated with the composite. This approach allows realizing an optical variant of an electrical strain gage rosette which can be exploited to resolve the general internal strain state of a composite material. A strain gage rosette usually consists of at least three strain sensors placed closely together under different angular orientations, allowing reconstructing the

strains relative to the material-symmetry axes regardless of the orientation of the sensor foil with respect to the composite. Knowing all strain components enables evaluating more accurately the strain-based composite failure criteria, leading to an improved mechanical design.^[4] Different rosette configurations can be selected. In this research, we have chosen a 45° rosette configuration. This is a configuration in which three sensors are used, and in which the second and third sensor are angularly displaced from the first sensor by 45° and 90°, respectively, see Figure 1a.

The sensor principle is analogous to that of FBG sensors, but instead of inscribing Bragg gratings in an optical fiber, they are inscribed in a polymer waveguide, forming a wavelength selective reflector. The Bragg grating sensors are implemented as periodic corrugations in the core of the waveguide. As a result, when exciting the waveguide with a broadband light source, a narrowband peak centered around a specific wavelength, called the Bragg wavelength λ_B , will be reflected. This wavelength is defined by the Bragg equation (for a first order grating, $\lambda_B = 2 \cdot n_{eff} \cdot \Lambda$, where n_{eff} is the effective refractive index of the propagating waveguide mode and Λ the grating pitch). Upon applying strain, both n_{eff} (due to the strain-optic effect) and the grating pitch change, leading to a shift in reflected wavelength. Therefore, tracking the Bragg wavelength shift (= sensor output signal) allows tracking the induced strain if the sensor sensitivity is known. The sensors are designed (see Supporting Information) to show reflection peaks in the wavelength range (1530-1590 nm) compatible with commercial interrogators which are also used to read out optical fiber sensors. All sensors are implemented in the same waveguide and therefore the grating pitch is chosen slightly differently so that their reflection peaks are separated in the wavelength domain and each sensor can be tracked independently. Figure 1b shows a typical reflection spectrum of 3 Bragg grating sensors with 502 nm, 506 nm, and 510 nm pitch realized in a single Ormocer®-based polymer waveguide.

Bragg gratings in silica optical fibers are sold commercially, and more recently Bragg gratings in polymer optical fibers are now starting to be considered for some applications.^[5]

Also, reports on Bragg gratings in planar polymer waveguides can be found, for realizing (tunable) lasers,^[6] (tunable) narrowband filters,^[7] temperature sensors,^[8] or strain sensors.^[9]

However, until now, those structures were never realized in thin foils suitable for multi-axial strain sensing or embedding in composites.

The grating corrugations are realized using UV nanoimprint lithography,^[10] and the waveguides either also using imprinting, or using laser direct-write lithography.^[11] The most important steps of the imprinting-based fabrication procedure are illustrated in Figure 1a and briefly discussed below; more details are given in the Supporting Information. First, a channel is created at the top surface of a spin-coated cladding layer using an imprinting stamp comprising the negative ridge-like patterns. After UV-curing this cladding layer, the core material is spin-coated on top of the imprinted channel and subsequently imprinted with another stamp comprising the grating corrugations. Finally, another cladding layer is spin-coated to obtain a symmetric cladding-core-cladding waveguide sensor stack. The (foil) substrate serves as an extra support to realize a stand-alone sensor, or can be removed after the fabrication to yield a minimally obtrusive, ultra-thin sensor which can be integrated in a mechanical structure to be monitored. Figure 1c shows a finalized sensor foil with an integrated 45° strain rosette, including a fiber connection for reading out the reflection spectrum.

The type of optical polymers used, determines the fabrication processes that can be applied, the resulting sensor properties (such as sensitivity), but also the mechanical properties and the interaction between the sensor foil and the mechanical structure to be monitored. For these reasons, this paper discusses the potential of the polymer sensor foils based on sensors realized using 2 types of materials: hybrid organic-inorganic materials, i.e. Ormocers®, and epoxy (EpoCore and EpoClad materials).^[12] Ormocer® has a relatively low optical loss at the

operating wavelengths and allows realizing long waveguides in which multiple Bragg grating sensors are integrated for multi-axial sensing. Using the epoxy, very thin sensor foils are realized which allows demonstrating the small bending radii and large elongations that these polymer foils can undergo. Furthermore, epoxy materials are widely used as matrices in composites, making this material ideal for realizing sensor foils compatible with embedding inside composites.

The response of the multi-axial sensor rosette to strain was determined by bending it under 2 orthogonal directions when mounted on a composite plate (see Experimental section). The plate ($25 \times 31 \text{ cm}^2$, 0.81 mm thick, composed by 2 layers of unidirectional glass fiber oriented at 0° degree and kept together by an epoxy-matrix) contained a surface-mounted multi-axial (MA) Ormocer® foil having 3 sensors oriented under 0, 45 and 90 degrees, as illustrated in Figure 2a. Additionally, 2 reference FBG sensors, which have a sensitivity of $1.2 \text{ pm}/\mu\epsilon$, were mounted under 0 and 90 degrees. The sensitivity of the Ormocer®-based sensors (i.e. $1.4 \text{ pm}/\mu\epsilon$, or 17% larger than for the FBG sensor) was determined using a separate tensile test (see Supporting Information). A controlled mechanical excitation of the sensors was applied with a standard 4-point bending (4PB) test procedure.^[13] In this condition, the strain acting on the sensors is uniform and linear with respect to the force and displacement imposed by the test bench (see Experimental Section), which facilitates analyzing the sensor behavior. Figure 2b shows the output signals (i.e. Bragg wavelength shift) of the sensor under test along with those of the reference FBG sensors as a function of applied displacement while loading the plate in tension and compression under 0° and 90° . This experiment clearly demonstrates the sensor's capability for multi-axial strain sensing: the sensor oriented along the direction of the largest strain, shows the largest signal, while the perpendicularly oriented sensor shows no or a small signal. It can be shown (see Supporting Information) that the theoretically expected strain at the surface and orthogonal to the bending direction is 0. This is reflected in the

signals from the FBG sensors, but not entirely in the signals from the polymer sensor foil which are clearly non-zero in that direction. The reason is the non-zero strain in the direction perpendicular to the foil which is translated into an in-plane strain component in the sensor through the Poisson effect. Because of the much lower Young's modulus and higher Poisson's coefficient compared to that of FBG sensors, this effect is only seen in the polymer sensor. Eventually, this effect can therefore be used to our advantage for recording the out of plane strain inside composite structures.

A third sensor (in the case of this experiment the sensor oriented under 45°) is required to retrieve the engineering shear strain γ ($\gamma = 0.5 \epsilon_{45} - \epsilon_0 - \epsilon_{90}$, where the subscripts indicate the sensor orientation). In our case, considering the orientation of the gratings, the laminate lay-up and the applied load, the shear strain has to be zero. Therefore, the strain measured by the sensor oriented at 45° must be equal to the semi-sum of the strains measured by the other two sensors which is in agreement with the results shown in Figure 2b.

Since there is a linear relation between strain and displacement, the results of this test demonstrate the linearity of the multi-axial sensor with respect to strain. Comparing the loading tests in tension and compression, it can be seen that all sensors show very similar (but inverse) behavior showing the same sensitivities for positive and negative values of strain. Comparing the loading tests under 0 and 90° , it can be seen that the sensitivity of the sensor MA- 0° (when loading is along 0° degrees direction) appears slightly higher than the sensitivity of sensor MA- 90° (when loading is along 90° direction), while the sensitivity of the reference FBG sensors is the same in both directions. The reason for this slight deviation is attributed to the accidental thicker glue layer under sensor MA- 0° situated close to the connector so that it locally undergoes a larger strain when bending. However, for the MA- 90° sensor, which did not suffer from this, the observed 29% higher sensitivity compared to the reference FBG sensors is very similar to the expected 17% difference in sensitivity. Figure 2c,

in which the plate was bent by imposing a periodic displacement according to a haversine function with a frequency of 0.5 Hz, further demonstrates the multi-axial sensor behavior as a function of time and the ability to synchronously readout the 3 sensors.

The previous shows the design flexibility of the photonic sensor foil technology: a large amount of sensors with a defined orientation can be multiplexed in a single foil. In addition, polymer sensor foils have many other attractive properties. Since polymers have a low Young's modulus and can be shaped into thin flexible foils, the resulting sensors can have a large deformability, or even conformability when stretchable materials are used.^[3] This capability was demonstrated by realizing polymer sensors without supporting foil carrier, to achieve the thinnest possible sensor foils. This was accomplished by fabricating the sensor stack on top of a release layer, so that free-standing ultra-thin sensor foils could be obtained after releasing (see Supporting Information). Using this technique, 35, 50 and 100 μm thick sensor foils were realized in optical epoxy materials (more precisely EpoCore and EpoClad from MicroResist Technology)^[12]. Although functional, the 35 μm thick foils were found to be too thin and fragile to handle them properly, while the 100 μm foils were already rather thick. Therefore, most of the work was done based on the 50 μm thick foils, being very thin, but still thick enough to provide sufficient mechanical stability for handling in most applications. The flexibility of the sensor and ability to undergo small curvature radii was demonstrated by subjecting a 50 μm thick sensor foil glued onto a 175 μm thick PET foil down to increasingly small bending radii (see Experimental Section). Figure 3a shows the Bragg wavelength shift of such a sensor as a function of the sensor radius of curvature and estimated strain. It can be seen that the sensor can be bent down to small radii (~ 11 mm) and can be strained significantly without compromising functionality. From a mechanical point of view, even smaller bending radii are possible, but will lead to increased optical bending loss in the optical waveguide and therefore reduced sensor reflection signal for the current design.

To further demonstrate the sensor's capabilities, the 50 μm thick sensor foil glued onto PET foil was slowly elongated using a tensile tester until the glue started delaminating. Figure 3b shows that this yielded a maximum Bragg wavelength shift of 18 nm, corresponding to a strain of 1.4%. As is commonly observed for Bragg grating sensors undergoing such elongations, spectral widening was visible as illustrated in the figure insets. Nevertheless, after releasing, the sensor spectrum recovered to the original state.

The mechanical flexibility and low thickness is also advantageous for unobtrusively integrating those sensor foils in mechanical structures. For the epoxy sensors, the integration in composite structures is particularly interesting, since a large class of composite materials is based on a similar epoxy resin keeping reinforcement fibers together. Therefore, it was investigated whether such sensors of different thickness survive a typical resin infusion composite fabrication cycle^[14] (see Experimental Section) when inserted inside the composite. The 35 μm thick sensor foils showed significant deformation due to their too low stiffness. The 50 and 100 μm thick sensor foils, however, showed only minor deformation as illustrated in Figure 3c, showing cross-sectional microscope images at different magnifications along the length and width of the rectangular sensor foils embedded in a composite specimen. Furthermore, there was no significant distortion observed in the sensor signal following the embedding process, except for a wavelength shift of a few nm caused by residual strain in the composite after cooling down, a phenomenon inherent to the fabrication technique.^[15] The correct operation of the sensor was verified by performing a tensile test on a composite specimen having a 50 μm thick sensor foil integrated. The obtained sensor signal as a function of applied strain (1.08 pm/ $\mu\epsilon$, see Figure 3d) was only slightly lower than the case in which such a sensor was realized on the surface of such a specimen (1.27 pm/ $\mu\epsilon$, see Supporting Information), the slight difference may be due to a difference in the strain transfer from the host material to the sensor.^[16]

In conclusion, a new class of thin polymer strain sensor patches has been realized. The various gains of using polymers and their implication on the resulting sensor properties have been shown. Polymer sensor foils can be flexible and thin, making them largely deformable and suitable for unobtrusively integrating in mechanical structures. By using polymers, a large design freedom is possible and depending on the concrete type of polymer, the sensors can be optimized for different aspects. In this work, it was demonstrated that usingOrmocer® materials, which have a relatively low optical loss and are compatible with imprinting, a multitude of sensors in different orientations can be multiplexed in the same foil, for example to realize multi-axial strain sensors. Those sensors were found to be very linear and have a slightly higher sensitivity (i.e. 1.4 pm/ $\mu\epsilon$) compared to standard FBG sensors. Using another material, epoxy, it was demonstrated that functional sensor foils down to a thickness of 50 μm could be realized. Those sensors were bent down to a radius of 11 mm, elongated by 1.4% and were embedded in epoxy-based composite materials without degradation of the sensor performance.

Experimental Section

4-point bending tests on Ormocer® sensors: To characterize the multi-axial behavior of the sensor foil, it was subjected to compression and tension, along 2 orthogonal directions, labeled 0° and 90°. Therefore, it was glued (using a thin layer of EA 9483 epoxy-based Loctite) in the center of a 0.81 mm thick glass fiber reinforced composite plate measuring 25 x 31 cm² with reinforcement fibers parallel to the longest plate direction (= 0° direction). As a reference, 2 standard FBG sensors were glued parallel to both the 0° and 90° direction using the standard procedure as advised by the provider. For the bending test, a 5800R Electro-mechanical Instron was used, equipped with fixtures having cylindrical rods with a center-to-center spacing of 228 mm below the plate and 143 mm on top of the plate, as illustrated in Figure 2a. To apply compression or tension on the sensors, the bottom fixture was moved up

with a speed of 0.1 mm/s and a maximum displacement of 5 mm to induce bending of the plate. In such a 4-point bending test, the displacement and resulting maximum strain at the center of the plate are linear with respect to the applied load. Details on the theoretically expected mechanical behavior based on a simplified model can be found in the Supporting Information.

Bending tests on epoxy sensors: The 50 μm thick epoxy sensors were tested towards small curvature radii and large elongations. To facilitate handling, a test specimen was made by gluing the ultra-thin sensor, having a fiber connected to it, onto a 175 μm thin PET foil strip using a thin layer of EA 9483 epoxy-based Loctite. This specimen was bent in a U-shape by clamping it between 2 parallel horizontal plates of which the separation was precisely controlled using a translation stage. By progressively moving the top plate down, an increasingly small bending radius was induced. A camera was used to visualize the bent specimen from the side and based on those images and post processing in Matlab, the radius of curvature at the location of the sensor was determined. From the radius, the theoretically expected strain in the sensor was estimated using the Euler–Bernoulli beam theory assuming uniform material properties.

Embedding epoxy sensor foils in composites: The composite test specimens were fabricated using the vacuum assisted resin infusion process.^[14] In this process, a stack of layers of dry fibers (i.e. reinforcement fibers) are impregnated with a low-viscosity resin (i.e. matrix) by means of a vacuum created inside a mold. Four layers of unidirectional glass fiber (500 g/m² UDO E S500 from SGL Group)^[17] were impregnated with a 2-component epoxy resin (RIMR135+RIMH137 from Hexion)^[18]. Epoxy sensor foils with a thickness of 35 μm , 50 μm and 100 μm , a width of 10 mm and length of 40 mm were placed between the middle two

fiber sheets. After impregnating the fibers, the resin was cured for 24 hours at room temperature and a post-cure was performed in an oven (15 hours at 80°C). For inspection of the embedding quality, cross-sections were made along the sensor width and length direction (Figure 3c). For testing the functionality of the embedded sensor, a composite test specimen was cut out (measuring $t \times w \times l = 1.5 \times 40 \times 110 \text{ mm}^3$), having a thin sensor foil integrated close to the short side of the specimen, as illustrated in the inset of Figure 3d. This allowed gluing an optical fiber to the specimen for reading out the sensor data. Afterwards, steel blocks were glued on both short sides of the specimen, it was clamped in an Instron® (model 8801) tensile tester, and actuated with a crosshead speed of 0.2 mm/min. A 10 mm gauge length dynamic extensometer with a strain range of 10 % was mounted on the specimen to record the actual strain at the grating sensor location and the resulting sensor signal (i.e. Bragg wavelength) was recorded as a function of strain.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

This work was conducted in the framework of the project Self-Sensing Composites (SSC), funded by the Flemish Government through the SBO project IWT-nr 120024. We also thank the Hercules foundation to provide funding for the APPLIE4MOS polymer prototyping line and for the e-beam system used in this research.

Received: ((will be filled in by the editorial staff))

Revised: ((will be filled in by the editorial staff))

Published online: ((will be filled in by the editorial staff))

References

- [1] G. Chiesura, A. Lamberti, Y. Yang, G. Luyckx, W. Van Paepegem, S. Vanlanduit, J. Vanfleteren, J. Degrieck, *Sensors* **2016**, 16, 866.
- [2] a) G. Luyckx, E. Voet, N. Lammens, J. Degrieck, *Sensors* **2011**, 11, 384; b) M. Majumder, T. K. Gangopadhyay, A. K. Chakraborty, K. Dasgupta, D. K. Bhattacharya, *Sens. Actuators, A* **2008**, 147, 150.
- [3] a) J. Hu, L. Li, H. Lin, P. Zhang, W. Zhou, Z. Ma, *Opt. Mater. Express* **2013**, 3, 1313; b) J. Missinne, S. Kalathimekkad, B. Van Hoe, E. Bosman, J. Vanfleteren, G. Van Steenberge, *Opt. Express* **2014**, 22, 4168.
- [4] a) C. T. Herakovich, *Mechanics of fibrous composites*, Wiley, Weinheim, Germany 1998; b) L. J. Hart-Smith, *Compos. Sci. Technol.* **1998**, 58, 1151.
- [5] W. Zeng, in *Handbook of Smart Textiles*, (Ed: X. Tao), Springer Singapore, Singapore **2015**, 109.
- [6] a) K.-J. Kim, J.-W. Kim, M.-C. Oh, Y.-O. Noh, H.-J. Lee, *Opt. Express* **2010**, 18, 8392; b) Y.-O. Noh, H.-J. Lee, J. J. Ju, M.-s. Kim, S. H. Oh, M.-C. Oh, *Opt. Express* **2008**, 16, 18194; c) S. Döring, M. Kolloosche, T. Rabe, J. Stumpe, G. Kofod, *Adv. Mater.* **2011**, 23, 4265.
- [7] a) B. Yun, G. Hu, Y. Cui, *Opt. Commun.* **2014**, 330, 113; b) A. Liu, Z. Zhang, D. d. Felipe, N. Keil, N. Grote, *IEEE Photonics Technol. Lett.* **2014**, 26, 313.
- [8] a) N. T. Benítez, J. Missinne, Y. Shi, G. Chiesura, G. Luyckx, J. Degrieck, G. V. Steenberge, *IEEE Photonics Technol. Lett.* **2016**, 28, 1150; b) S. Sherman, H. Zappe, *Proc. Technol.* **2014**, 15, 702.
- [9] a) M. Rosenberger, S. Hessler, S. Belle, B. Schmauss, R. Hellmann, *Opt. Express* **2014**, 22, 5483; b) M. Rosenberger, B. Schmauss, R. Hellmann, *Sensors* **2017**, 17, 1964.
- [10] C. Zhang, H. Subbaraman, Q. Li, Z. Pan, J. G. Ok, T. Ling, C.-J. Chung, X. Zhang, X. Lin, R. T. Chen, L. J. Guo, *J. Mater. Chem. C* **2016**, 4, 5133.
- [11] A. Elmogi, E. Bosman, J. Missinne, G. Van Steenberge, *Opt. Mater.* **2016**, 52, 26.
- [12] Microresist Technology, <http://www.microresist.de>
- [13] ASTM standard.D6272 - *Standard Test Method for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials by Four-Point Bending*.
- [14] J. Summerscales, T. J. Searle, *Proc. Inst. Mech. Eng., Part L* **2005**, 219, 45.
- [15] N. Lammens, D. Kinet, K. Chah, G. Luyckx, C. Caucheteur, J. Degrieck, P. Mégret, *Composites, Part A* **2013**, 52, 38.
- [16] a) G. Luyckx, E. Voet, W. D. Waele, J. Degrieck, *Smart Mater. Struct.* **2010**, 19, 105017; b) E. Voet, G. Luyckx, W. D. Waele, J. Degrieck, *Smart Mater. Struct.* **2010**, 19, 105018.
- [17] SGL Group, <http://www.sglgroup.com/>
- [18] Hexion, <http://www.hexion.com/>

Figure 1. (a) Imprinting-based fabrication process for realizing thin sensor foils with 3 grating sensors (having pitches Λ_1 , Λ_2 , Λ_3) all multiplexed in the same optical waveguide. (b) Typical reflection spectrum of the sensor foil. (c) Realized sensor foil illustrating the grating locations (overlay drawing) and showing the fiber connector for readout.

Figure 2. Multi-axial (MA) Ormocer® sensor foils (a) Four point bending test setup including the sensor configuration as seen from the top. (b) Sensor foil output signals (= Bragg wavelength shift) compared to the output signals from reference FBG sensors for loading of the plate in compression and tension and along two orthogonal directions. (c) Sensor output signals as a function of time when cyclically compressing along the 3 directions under which the sensors are oriented.

Figure 3. Ultra-thin epoxy sensor foils: (a) Sensor signal as a function of estimated radius of curvature and corresponding strain. (b) Signal of a sensor undergoing a large elongation tensile test. The insets show the reflection spectrum at different instants during the test (c) Cross-sectional images (along and perpendicular to the reinforcement fibers) of 50 and 100 micron thick sensor foils embedded in a composite (d) Sensor signal as a function of applied strain for a 50 micron thick foil embedded in a composite test specimen.

Table of contents entry

Thin and flexible photonic sensor foils allow multi-axial strain sensing of composites by multiplexing a multitude of Bragg grating sensors. Those foils can be made very thin (down to 50 μm) making them suitable for embedding. Owing to their low Young's modulus and flexibility, polymer sensor foils can be bent to small curvature radii and withstand large elongations.

Keywords

Bragg gratings, composites, foil, multi-axial strain sensors, polymer waveguides.

J. Missinne*, N. Teigell Benítez, A. Lamberti, G. Chiesura, G. Luyckx, M-A. Mattelin, W. Van Paepegem, G. Van Steenberge

Thin and flexible polymer photonic sensor foils for monitoring composite structures

Supporting Information

Thin and flexible polymer photonic sensor foils for monitoring composite structures

Jeroen Missinne, Nuria Teigell Benítez, Alfredo Lamberti, Gabriele Chiesura, Geert Luyckx, Marie-Aline Mattelin, Wim Van Paepegem, Geert Van Steenberge*

1.Ormocer®-based multi-axial sensor foils**A. Multiplexed Bragg grating design**

The reflection wavelength or so-called Bragg wavelength λ_B is defined by the Bragg equation, i.e. $\lambda_B = 2 \cdot n_{\text{eff}} \cdot \Lambda$. Since n_{eff} , the effective refractive index of the waveguide mode, mainly depends on the used waveguide materials, Λ (grating pitch) is the main design parameter that can be chosen to tune the Bragg wavelength. If multiple grating sensors (each having a Bragg wavelength $\lambda_{B,i}$) are implemented along the same waveguide, their grating pitch needs to be chosen slightly differently to yield individually discernable reflection peaks. Herein, a Bragg wavelength separation of about 12.5 nm was chosen. Since most commercially available Bragg grating readout systems operate roughly in the 1510-1590 nm wavelength band, the following grating sensor parameters were selected:

- Sensor 1, $\lambda_{B,1} \approx 1555$ nm, $\Lambda_1 = 510$ nm
- Sensor 2, $\lambda_{B,2} \approx 1542.5$ nm, $\Lambda_2 = 506$ nm
- Sensor 3, $\lambda_{B,3} \approx 1530$ nm, $\Lambda_3 = 502$ nm

Furthermore, the grating profile and grating length determine the shape of the reflection spectrum. A rectangular grating with a depth of 100 nm and a duty cycle of 50 % is chosen as a compromise between ease of fabrication through imprinting and optical properties. To be able to detect the Bragg wavelength shifts accurately, the reflectivity of the grating should be

as high as possible and the bandwidth as small as possible. An approximation of the spectral reflectivity $R(\lambda)$ response can be obtained using the coupled-mode theory [1]:

$$R(\lambda) = \left| \frac{j\kappa \sin(\sqrt{\delta^2 - \kappa^2} L)}{\sqrt{\delta^2 - \kappa^2} \cos(\sqrt{\delta^2 - \kappa^2} L) - j\delta \sin(\sqrt{\delta^2 - \kappa^2} L)} \right|^2$$

where

$$\delta = n_{eff} \left(\frac{2\pi}{\lambda} - \frac{2\pi}{\lambda_0} \right),$$

$$\kappa = \frac{2\pi^2 a^3}{3\lambda_B l d^3} \frac{n_{core}^2 - n_{clad}^2}{n_{core}} \left[1 + \frac{3\lambda_B}{2a\pi(n_{core}^2 - n_{clad}^2)^{0.5}} + \frac{3\lambda_B^2}{4a^2\pi^2(n_{core}^2 - n_{clad}^2)} \right],$$

a the grating depth, l the grating order (= 1 in our case), d the waveguide width (3 μm), L the grating length, and n_{core} , n_{clad} , n_{eff} the core refractive index, cladding refractive index and waveguide effective index respectively.

Figure S 1. 3 dB bandwidth (BW) and peak reflectivity of the rectangular Bragg grating in a single mode polymer waveguide as a function of the number of grating periods.

Simulations show (**Figure S 1**) that the bandwidth decreases and the peak reflectivity increases with increasing number of periods. However, longer gratings tend to show increased peak broadening due to the increasingly higher reflectivity of detuned wavelengths and due to grating pitch non-uniformity because of fabrication inaccuracy and furthermore short gratings are preferred for localized sensing. A grating with more than 5000 periods does not show a

significant further increase in reflectivity or decrease in bandwidth. Therefore, the grating sensor farthest from the fiber connector (sensor 3, see also **Figure S 2**) was implemented with 5000 periods. Although the intrinsic reflectivity of this grating is close to 100%, the waveguide propagation losses, bend losses and connection losses will reduce this value. The number of periods for the second and third grating was chosen so that the effective reflectivity of the 3 gratings does not differ too much so that the sensors are compatible with the limited dynamic range of commercial interrogators. Based on an estimated value for the propagation loss and bend loss, 4000 and 2800 periods were chosen for sensor 2 and 1 respectively. As can be seen from Figure 1c, the losses were slightly underestimated, but a difference <3 dB in effective reflectivity between the neighboring peaks was achieved, which allows the readout of the three peaks simultaneously.

The waveguide, in which the grating was implemented, was designed to be single mode. Since the cladding and core material refractive index (at $\lambda \approx 1550$ nm) are fixed (OrmoClad, RI = 1.528, OrmoCore, RI = 1.537), the cross-sectional dimensions of the waveguide (i.e. $3 \times 4 \mu\text{m}^2$) were chosen to obtain a mode field diameter close to that of a single mode fiber (to reduce the coupling losses) while at the same time satisfying the single mode condition. In this work, standard single mode fibers were coupled to the sensor foils because then they become directly compatible with commercial interrogators for their readout.

Figure S 2. Schematic layout of the multi-axial sensor showing the orientation of the 3 grating sensors and the location of the fiber connector.

To implement a strain rosette, the 3 sensors were oriented 45° with respect to each other, as illustrated in **Figure S 2**. In this research, the straight waveguide sections (where the gratings are implemented) were chosen relatively long (10 mm) to have enough margin for testing out various grating designs. This leads to a total sensor area of $15 \times 15 \text{ mm}^2$ (dashed gray box in the figure). However, since the final grating length of the longest grating (farthest from the coupling point) was chosen to be only 2.5 mm, the sensor patch can be made much more compact in the future and a total sensor area below $10 \times 10 \text{ mm}^2$ can be obtained.

B. Fabrication based on imprinting

Figure S 3. Schematic illustration of the waveguide + grating fabrication process based on imprinting.

TheOrmocer® sensors were fabricated using an imprinting process both to define the waveguides and the gratings as schematically illustrated in **Figure S 3**. The basic principle is that after a master mold is fabricated once, replica can be made in a transparent flexible polymeric working stamp, and each of them can then be used multiple times to imprint a UV-curable polymer resin yielding the same features as on the master after UV-curing. Once such working stamps were available, the sensors were fabricated as follows. First, a 18 μm thick layer of OrmoClad (a type of Ormocer® formulation) was spin-coated on a 175 μm thick PET foil as substrate (a) and using imprinting with a suitable working stamp, small channels were formed in this layer after UV-curing the OrmoClad (b,c,d). Then, a thin layer of diluted OrmoCore (another type of Ormocer® formulation with a higher refractive index) was spin-coated on top of the channels and after a baking step the layer was covered with a working stamp (e) containing the grating structures and was subsequently UV-cured (f). The use of the working stamp in this step has 2 functions: (i) to define the grating structures at the top

interface of the waveguide and (ii) to force the spin-coated OrmoCore material into the channels by capillary action. A final spin-coated OrmoClad layer finishes the fabrication process. Several standard baking steps were applied during this process, but were omitted in the description above for brevity.

Further details on the master mold and working stamp fabrication are given below.

After fabrication, a testable sensor foil was obtained by cutting away the unused edges of the PET substrate using a wafer dicer equipped with a diamond-coated blade. This step also exposes the waveguide cross-section so that a single mode fiber can be connected. This connectorization process is described below.

Master mold fabrication

The waveguide master mold was fabricated using standard mask lithography on a 3 μm thick EpoCore layer spin-coated on a 4" Si wafer substrate. After the developing step, the remaining EpoCore structures consisted of the reverse of waveguide ridges (i.e. channels) being 4 μm wide and 3 μm high on the Si wafer.

The grating master mold containing three gratings oriented 45° with respect to each other was fabricated using e-beam lithography on a 100 nm thick PMMA layer (950-PMMA, A3 version; dose $350 \mu\text{C cm}^{-2}$) coated on a 4" Si wafer substrate. After exposure, the sample was developed for 30 s in a MIBK developer : IPA mixture (in a 1 : 3 ratio) followed by a 30 s rinsing step in IPA.

Working stamp fabrication

Figure S 4. Schematic illustration of the working stamp fabrication process starting from a master mold.

As illustrated in **Figure S 4**, the master mold (a) was replicated in an PFPE flexible stamp material by first casting a liquid mixture of a perfluoropolyether (PFPE) polymer (Solvay Fomblin® MD40) and photoinitiator (Irgacure) in a 1 : 0.03 ratio by weight onto the master after which it was distributed over the wafer by spin-coating at a slow speed (500 rpm) (b). Then, the sticky side of a transparent PET-based adhesive foil was brought in contact with the liquid layer (c) and the PFPE layer was UV cured with a broadband UV source (30 s at 35 mW/cm^2), after which the created flexible mold was released from the master mold yielding the so-called working stamp.

The working stamp with the grating rosette was performed using an analogous process flow.

With this process, 2 working stamps were fabricated, one with the waveguide structures and another with the grating structures.

2. Epoxy-based ultra-thin sensor foils

A. Design of the Bragg grating sensors

The epoxy single mode waveguides consist of an EpoCore (refractive index 1.577 at $\lambda = 1550 \text{ nm}$) waveguide core surrounded by an EpoClad (refractive index 1.570 at $\lambda = 1550 \text{ nm}$)

cladding. Since the refractive index difference between both is slightly different than for the Ormocer® materials used, the cross-sectional waveguide dimensions were also chosen differently, i.e. $5\text{ }\mu\text{m} \times 5\text{ }\mu\text{m}$. These dimensions were optimized previously to yield waveguides with low coupling loss to standard telecom single mode fibers [2].

The grating pitch was 505 nm, yielding a Bragg wavelength of about 1573 nm, which is compatible with available readout equipment.

B. Fabrication using laser-direct writing

Figure S 5. Schematic illustration of the epoxy waveguide fabrication process.

Since the final aim was to achieve ultra-thin sensor foils, the sensors were fabricated on a temporary glass carrier with release coating (PI 2611 polyimide from HD microsystems) so that the epoxy sensor layers could be released afterwards. In such a way the final sensor foil thickness depends on the total spin-coated layer thicknesses. Herein, sensor foil thicknesses of $35\text{ }\mu\text{m}$, $50\text{ }\mu\text{m}$ and $100\text{ }\mu\text{m}$ were realized.

The fabrication process is schematically illustrated in **Figure S 5** and described below. First, an EpoClad layer with half the final sensor thickness was spin-coated on the glass substrate with release coating. The adhesion of the EpoClad onto the release layer was good enough for the temporary adherence during the fabrication process, but still weak enough so that it can be released afterwards. Then, a grating was imprinted in the spin coated layer (a), similarly as described above. However, since the EpoClad coating is solid at room temperature, the imprinting was performed on a hotplate at elevated temperature. Afterwards, a 5 μm thick EpoCore layer was spin-coated (b) which was selectively exposed to UV using a Heidelberg DWL66+ direct-write lithography system (c). After developing the unexposed material (d), a second EpoClad layer with half the final foil thickness was applied. Using this technology, several sensor foils could be fabricated on the same temporary carrier and were afterwards cut out to the required dimensions using a Nd:YAG laser ($\lambda = 355 \text{ nm}$). After the cutting step, the sensor foils could easily be peeled off due to the limited adhesion of the release coating used.

3. Calibration of the sensors: sensitivity analysis of single sensors

In order to calibrate the sensor's sensitivity to strain and temperature, simple test specimens were made consisting of epoxy-based andOrmocer®-based sensors fabricated on top of a glass fiber reinforced composite (FR4) substrate. The fabrication steps were very similar to those which were used for the final sensors which are described above. The cross-sectional dimensions of the waveguides were 4 μm x 3 μm and 5 μm x 5 μm for the Ormocer® and the epoxy sensors respectively, which are the same dimensions than those of the final sensors.

A. Preparation of the specimen for testing

To make a testable and robust sample, an SMF-28 single mode optical fiber was pigtailed to the sensor waveguide using a UV-curable epoxy and a dedicated connector. In addition, to clamp the sample into the tensile tester, steel spacers were glued on both extremes of the specimen (see Figure 3d).

B. Sensor sensitivity with respect to strain

Tensile tests were performed in displacement control with a crosshead speed of 0.2 mm min^{-1} using an 8801 servo-hydraulic test machine from Instron® with a 100 kN load cell. A 10 mm gauge length dynamic extensometer with a strain range of $\pm 10\%$ was mounted on the specimen (at the grating location) to track the deformation and strain.

The reflection spectrum peak was tracked using a dedicated optical fiber sensor interrogator (FBG-Scan804 interrogator from FBGS Technologies GmbH) operating from 1510 nm to 1590 nm and with a resolution of 1 pm.

Since the data acquisition from the extensometer and the optical fiber sensor interrogator were performed at different rates, the acquisition time stamp was used for synchronization. The strain data was linearly fitted with respect to time and the result of this linear fit was used to correlate the recorded strain with the recorded Bragg wavelength.

The data obtained from the tensile tests is shown in **Figure S 6**. The results given by the associated linear fits lead to sensitivities of $\Delta\lambda/\Delta\epsilon = 1.27 \text{ pm}/\mu\epsilon$ for the epoxy-based and $\Delta\lambda/\Delta\epsilon = 1.41 \text{ pm}/\mu\epsilon$ for theOrmocer®-based sensors. These values are comparable (or slightly higher) than those previously reported for unetched polymer optical fiber sensors [3][4]. Commercial DTG®-type silica-based fiber sensors have a typical sensitivity of $1.2 \text{ pm}/\mu\epsilon$ when operating in the same wavelength range.

Figure S 6. Recorded Bragg wavelength as a function of recorded strain for (left) epoxy and (right) Ormocer®-based polymer waveguide Bragg grating sensors.

C. Sensor sensitivity with respect to temperature

The temperature tests were performed in a temperature controlled environmental chamber (Eurotherm2408 from Instron®) which has a port on the bottom to allow the pigtailed fiber to be connected to an interrogator outside the chamber. The sample was positioned in the center of the chamber with a Type K thermocouple measuring the environmental temperature in the near vicinity of the grating. The temperature of the chamber was ramped up between 20 °C and 60 °C. The sensor data acquisition was performed (with the same interrogator previously used) every 5 °C after letting the temperature stabilize in the chamber for 5 minutes.

The results of the temperature tests are shown in **Figure S 7** and the sensitivities given by the associated linear fits are $\Delta\lambda/\Delta T = -90$ pm/°C for the epoxy-based and $\Delta\lambda/\Delta T = -250$ pm/°C for the Ormocer®-based sensors.

Figure S 7. Recorded Bragg wavelength as a function of recorded chamber temperature for (left) epoxy and (right) Ormocer® based polymer waveguide Bragg grating sensors.

4. Theoretical analysis of a composite plate undergoing a 4-point bending test

Figure S 8. Illustration of the plate undergoing the 4 point bending configuration.

According to classical laminate plate theory CLPT [5], the bending moment per unit width acting on the edge with normal $x=1$ is given by

$$M_1 = -D_{11} \frac{\partial^2 w}{\partial x^2} - D_{12} \frac{\partial^2 w}{\partial y^2} - D_{16} \frac{\partial^2 w}{\partial x \partial y} \quad (1)$$

where D_{11} , D_{12} and D_{16} are the bending stiffnesses and w the out of plane displacement.

Since the glass reinforcement fibers are aligned with the x axis, $D_{16}=0$. Furthermore, for a laminate composed by two unidirectional layers of parallel reinforcement fibers at 0 degree, D_{11} and D_{12} are given by

$$D_{11} = \frac{E_1 h^3}{12(1 - \nu_{12} \nu_{21})} \quad (2a)$$

$$D_{12} = \frac{E_2 h^3 \nu_{12}}{12(1 - \nu_{12} \nu_{21})} \quad (2b)$$

For a 4PB test, the displacement enforced by the test bench will impose a cylindrical deformation of the plate ($\frac{\partial^2 w}{\partial y^2} = 0$). The bending moment at $x=0$ and $x=l$ can be written as follows

$$M_1(x = 0, x = l) = \frac{F(L - l)}{4b} = -D_{11} \frac{\partial^2 w}{\partial x^2} \quad (3)$$

By integrating Equation (3) with respect to x and considering that at $x=0$ & $x=L$, the out-of-plane displacement w is equal to displacement imposed by the test frame w_f ($w(x=0$ & $x=L)=w_f$), the following expression for $w(x)$ is obtained:

$$w(x) = -\frac{F(L - l)}{8bD_{11}} x(x - l) + w_f \quad (4)$$

Using CLPT, the axial strains acting along the directions parallel (0 degree) to the reinforcement fibers can be derived from equation (4) as follows

$$\varepsilon_0(x, z) = \frac{\partial u}{\partial x} + \frac{1}{2} \left(\frac{\partial w}{\partial x} \right)^2 - z \frac{\partial^2 w}{\partial x^2} = \frac{\partial u}{\partial x} - \frac{1}{2} \left[\frac{F(L - l)}{8bD_{11}} (2x - l) \right]^2 + z \frac{F(L - l)}{4bD_{11}} \quad (5)$$

where u is the midplane displacement. Since at $x=0$ & $x=l$ the simply-supported boundary condition is imposed, u can be written as:

$$u(x) = \sum_{m=1,3,5,\dots}^{\infty} u_m \cos\left(\frac{m\pi x}{l}\right) \quad (6)$$

and hence

$$\frac{\partial u(x)}{\partial x} = \sum_{m=1,3,5,\dots}^{\infty} \frac{-\pi m}{l} u_m \sin\left(\frac{m\pi x}{l}\right) \quad (7)$$

At the location of the sensor foil ($x=l/2, z=h/2$), $\frac{\partial u(x)}{x} = 0$ and therefore equation (5) evaluates in:

$$\varepsilon_0(x = l/2, z = h/2) = \frac{Fh(L - l)}{8bD_{11}} \quad (8)$$

The strain ε_{90} acting along the direction orthogonal (90 degree) to the reinforcement fibers is negligibly small in the 4-point bending configuration considered [6].

Furthermore, considering the loading condition, the laminate layup and the orientation of the sensor foil with respect to the material symmetry axis, no in plane shear strain is present. Consequently, given the 45 degree rosette configuration of the sensors, the axial strain measured by the grating in the 45 degree must verify the following relation [7]:

$$\varepsilon_{45} = \frac{\varepsilon_0 + \varepsilon_{90}}{2} \quad (9)$$

5. Additional measurements

A. Ultra-thin epoxy sensor, bending, in tension

The 50 μm ultra-thin epoxy sensors were subjected to a number of tests to demonstrate their potential. In the main document, it was already shown that the sensor foil can be bent to small radii, can undergo large strains and can be embedded in composite materials without compromising performance. Here, the results of an additional cantilever bending test are discussed.

A test specimen was made by gluing the ultra-thin sensor, having a fiber connected to it, onto a 175 μm thin PET foil strip. This specimen was clamped on 1 side with the sensor facing upward, as illustrated in **Figure S 9a**. In this way, the sensor is loaded in tension when pushing down the specimen at the free end using a pin. By progressively moving the pin down, an increasingly large deflection of the specimen and hence tension in the sensor was induced. A camera was used to image the bent specimen from the side and based on those images and post processing in Matlab, the bending radius at the location of the sensor was estimated. From the bending radius, the theoretically expected strain in the sensor was estimated using

the Euler–Bernoulli beam theory assuming uniform material properties. The results are plotted in **Figure S 9b**, showing the recorded Bragg wavelength shift as a function of strain and radius of curvatures of the test specimen

Figure S 9. (a) Side view of the test specimen bent using a pin during the cantilever test and (b) resulting sensor output (Bragg wavelength shift) as a function of strain in the sensor and radius of curvature of the test specimen.

References

- [1] Yariv, A. (1973). Coupled-mode theory for guided-wave optics. *IEEE Journal of Quantum Electronics*, 9(9), 919-933.
- [2] Elmogi, A., Bosman, E., Missinne, J., & Van Steenberge, G. (2016). Comparison of epoxy-and siloxane-based single-mode optical waveguides defined by direct-write lithography. *Optical Materials*, 52, 26-31.
- [3] Chen, X., Zhang, C., Webb, D. J., Peng, G. D., & Kalli, K. (2010). Bragg grating in a polymer optical fibre for strain, bend and temperature sensing. *Measurement Science and Technology*, 21(9), 094005.
- [4] Rajan, G., Ramakrishnan, M., Semenova, Y., Ambikairajah, E., Farrell, G., & Peng, G. D. (2014). Experimental study and analysis of a polymer fiber Bragg grating embedded in a composite material. *Journal of lightwave technology*, 32(9), 1726-1733.
- [5] Reddy, J. N. (2004). *Mechanics of laminated composite plates and shells: theory and analysis*. CRC press.
- [6] Nimbolkar, P. V., & Jain, I. M. (2015). Cylindrical bending of elastic plates. *Procedia Materials Science*, 10, 793-802.
- [7] Pendleton, R., & Tuttle, M. (Eds.). (2012). *Manual on experimental methods for mechanical testing of composites*. Springer Science & Business Media.