

5. Conclusions and perspectives

The most important objective of this thesis was the analysis of one type of mesoporous silica films representing a very suitable low- n substrate for waveguide structures. It has been demonstrated that these optically perfect MSFs can be fabricated in a sol-gel process by dip-coating in conditioned air.

It has been found that the chemical composition of the precursor solution does not solely determine the structure of the synthesis product. While some previous investigations of Pluronic-based syntheses [34, 53, 54] have neglected the processing factors, this work shows the dramatic influence of processing conditions on the film structure and its properties.

Sol-gel processes lead to systems which are frozen in a non-equilibrium state, which is highly dependent on the exact reaction pathway and, therefore, on the used processing conditions such as humidity, drawing rates, diffusion paths, and heating and cooling rates. Non-equilibrium structures have been reproducibly stabilized in the regarded mesoporous films. If the processing conditions are kept constant in defined ranges, the synthesized structures have constant properties which significantly differ from the equilibrium phases.

The humidity during film fabrication turned out to be a decisive processing parameter for the fabrication of optically perfect films. In the relative humidity range between 20 and 40% the desired A-type films have been reproducibly synthesized. The control of the film thickness in the range of $d = 300\text{-}1100$ nm, which is important for many applications, is possible via the drawing velocity v .

The coating with a tilting movement is a suitable method to fabricate films with a thickness gradient. Such samples are useful for those experiments, in which the controlled variation of the thickness is an important parameter, because it reduces the number of samples with different thicknesses. An exponent $\alpha = 0.637 \pm 0.044$ has been found for the expression $d = \text{const} \cdot v^\alpha$, which is very close to $2/3$, predicted by the Landau-Levich theory [49].

The nanostructure of the films has been determined using different characterization methods like small angle X-ray scattering (SAXS), transmission electron microscopy (TEM), and atomic force microscopy (AFM). A-type films have a stable wormlike structure with cylinder-like channels. The channels are randomly oriented, but have uniform, well-defined pore distance of about 8 nm. This is characteristic for dense or nearly dense packed channel systems of the SBA-15-like synthesis class [34].

The thickness of the films can be controlled steadily up to 1 μm which is sufficient for most waveguiding structures. This requirement was not reached by other syntheses [53] and, therefore, the applications of those films are excluded.

A-type films turned out to have most useful properties for optical applications. They exhibit an ultra-low refractive index of $n = 1.18 \pm 0.01$, low birefringence (below 0.01), extremely flat surface (RMS = 0.5 nm), sufficient mechanical and chemical stability, and low optical scattering. The high porosity of $(59 \pm 2)\%$ is the reason for the low refractive index of the A-type films.

This structure can resist the stress induced by further treatments. The films are thermally stable up to about 950°C . Therefore, they can be used in many further processing techniques. Due to these extraordinary properties, A-type films are interesting for applications in integrated optical structures. They are well suited as low- n supports for waveguides.

A novel calcinable lamellar structure has been found for the B-type films at processing conditions of relative humidity above 45%. This structure consists of layers with an interlayer spacing of 3 nm and a sustaining network with a typical mesh width of 20 nm. The layers were also made visible in the TEM and AFM images and the determined spacing between them was in very good agreement with the SAXS results. The proposal of the sustaining network was made on the basis of AFM phase images in consideration of the energy dissipation. Such structures may

have interesting applications in biocatalysis and drug release since their usable pore size is very large.

Waveguides on mesoporous substrates have a good mechanical and thermal stability and allow integration into lithographic structuring techniques. In the joint effort with the cooperation partners in IPHT Jena and TU Hamburg-Harburg a 2D photonic crystal line defect resonator made of a P(MMA-DR1) polymer was realized. The resonator structure on the mesoporous substrate showed a high transmission in the resonance peak. The high vertical index contrast improves the transmission of the device significantly. Due to ultra-low refractive index substrate MSF, no radiation losses were observed on the air band side of the photonic band gap.

MSFs might also be a suitable support for the deposition of the ferroelectric material PZT. The crack formation in the PZT films was significantly suppressed when they were deposited directly on the MSF. However, further investigations are needed in order to exclude possible changes of the MSF support.

Besides fabricated PZT films, first attempts for the PZT structuring have been carried out. A sol-gel method offers an alternative to the conventional lithographic structuring of photonic crystals. However, the extension of inverse opals to thin films remains an open challenge on the way to the realization of a switchable PhC.

The mesoporous silica films have proven to be compatible with many waveguide core materials [115] and can be regarded as a general approach to 2D PhC slab waveguides when a low refractive index support is required. As already noticed, low- n supports have three important advantages: one can avoid working with leaky modes, the penetration depth into the substrate is lower, and the system is nearly symmetric. With the use of a low- n cover layer the system could be made fully symmetric. The symmetry leads to decoupling of TE and TM modes, which is also very useful for an easier design of functional waveguide structures or structures using non-linear optic (NLO) effects. Perspectives for the use of MSFs in other systems which require low- n supports to achieve high refractive index contrast, e.g. nanowires as nanoscale light emitters, are also very promising [168].