Combining one and two photon polymerization for accelerated high performance (3 + 1)D photonic integration

Abstract: Dense and efficient circuits with component sizes approaching the physical limit is the hallmark of high performance integration. Ultimately, these features and their pursuit enabled the multi-decade lasting exponential increase of components on integrated electronic chips according to Moore’s law, which culminated with the high performance electronics we know today. However, current fabrication technology is mostly constrained to 2D lithography, and thermal energy dissipation induced by switching electronic signal lines presents a fundamental challenge for truly 3D electronic integration. Photonics reduces this problem, and 3D photonic integration is therefore a highly sought after technology that strongly gains in relevance due to the need for scalable application-specific integrated circuits for neural networks. Direct laser writing of a photoresin is a promising high-resolution and complementary metal-oxide-semiconductor (CMOS) compatible tool for 3D photonic integration. Here, we combine one and two-photon polymerization (TPP) for waveguide integration for the first time, dramatically accelerating the fabrication process and increasing optical confinement. 3D additive printing is based on femtosecond TPP, while blanket irradiation with a UV lamp induces one-photon polymerization (OPP) throughout the entire 3D chip. We locally and dynamically adjust writing conditions to implement (3 + 1)D flash-TPP: waveguide cores are printed with a small distance between neighboring writing voxels to ensure smooth interfaces, mechanical support structures are printed at maximal distance between the voxels to speed up the process. Finally, the entire chip’s passive volume not part of waveguide cores or mechanical support is polymerized in a single instance by UV blanket irradiation. This decouples fabrication time from the passive volume’s size. We succeed in printing vertical single-mode waveguides of 6 mm length that reach numerical apertures up to NA = 0.16. Noteworthy, we achieve exceptionally low ~0.26 dB injection losses and very low propagation losses of ~1.36 dB/mm at $\lambda_0 = 660$ nm, which is within one order of magnitude of standard integrated silicon photonics. Finally, the optical performance of our waveguides does not deteriorate for at least ~3000 h after printing, and remains stable during ~600 h of continuous operation with 0.25 mW injected light.

Keywords: (3 + 1)D flash-printing; one-photon polymerization (OPP); single-mode waveguides; two-photon polymerization (TPP); ultra-fast 3D additive manufacturing.

1 Introduction

Additive manufacturing based on 3D printing is an innovative tool to create complex 3D devices. Direct laser writing (DLW) associated with two-photon polymerization (TPP) allows the creation of micron to sub-micrometer three-dimensional (3D) structures in diverse fields, such as micromechanical systems [1], microrobotics components [2] or biosciences [3]. In DLW, a tightly focused laser spot is translated through a photoresin to form a solid inside the TPP process’s reactive volume, which results in a sub-micron voxel [4]. In photonics, the DLW-TPP technique has been used to fabricate free-form and transformational components [5–7], point-to-point photonic wire-bondings [8], waveguides [9], spatial-filters [10], graded-index lenses [11] and photonic components [12, 13]. Simultaneously, this technique is advantageous for integrated photonic circuits [14, 15] due to its ability to locally and dynamically modify optical properties on feature sizes below the Abbe resolution limit.
Photonic waveguides are of fundamental importance for many optical components and to realize their implementation in day-to-day technologies [8, 16]. The ability of additively integrating photonic circuits with micron feature sizes opens new perspectives for applications, such as the scalability of integrated photonic neural network circuits [10, 17], chip-to-chip interconnects [18] or in building blocks for photonic waveguide integration in 3D [19, 20]. Micromachining of photonic waveguides by DLW-TPP is currently explored in diverse materials, i.e. glasses, crystals and polymers [21]. For additive DLW, the working distance of the microscope objective does not constrain the fabrication process and allows single-step printing on different substrates. The technique is therefore an excellent candidate for manufacturing complex and large integrated photonic circuits directly on top of other, e.g. semiconductor substrates, and generally promises to be highly CMOS compatible.

In standard photonic waveguides, light confinement is caused by total internal reflection at the interface between core and cladding. The main condition for this is that the refractive index of the core (n_{core}) is larger than the one of the cladding (n_{cladding}). For polymer waveguides in which the surrounding media is air, the core-cladding refractive index difference $\Delta n = n_{core} - n_{cladding} \approx 0.5$ is large and easily leads to multi-mode propagation at the feature sizes achievable with TPP for visible or near infrared (NIR) wavelengths [22]. Single-mode propagation can be ensured by a core diameter below the single-mode cut-off diameter. For $\Delta n = 0.5$, this is out of reach of current single-step additive DLW techniques. However, single-mode waveguides are fundamentally important and are broadly employed in integrated photonics, low-loss fibers [23] or for quantum and optical communications [24, 25]. To overcome this limitation, we have recently demonstrated a (3+1)D printing approach, where we leveraged precise control of the local degree of TPP polymerization, and in turn over $\Delta n$ on the level required for single-mode waveguides with $\mu$m core diameters [9].

In this article, we significantly advance this (3+1)D fabrication methodology, here demonstrated with the commercially available IP-S photosensitizer [26]. In order to accelerate fabrication while simultaneously improving waveguiding performance, we combine three concepts dedicated to fabricating three essential parts of a circuit: (i) the waveguide cores, (ii) the waveguide cladding and (iii) mechanical support ensuring the stability of the 3D integrated circuit. Waveguide cores require $\mu$m resolution and smooth interfaces with their cladding to ensure low-loss propagation. We therefore use TPP with a fine resolution in the $(x, y)$-plane, i.e. a small hatching distance $h$, and a carefully optimized TPP writing power. Areas only acting as mechanical support do not require smooth surfaces and we use large TPP-writing powers and large hatching distances in order to reduce fabrication time. Finally, the majority of an integrated photonic circuit comprises material with a uniform refractive index lower than the one of waveguide cores. This, we achieve in a single-shot for the entire circuit via blanket ultraviolet (UV) irradiation with a controlled exposure dosage to precisely control $\Delta n$. Furthermore, the UV exposed regions also enhance the circuit's mechanical stability for small diameter waveguides and for waveguides having considerable lengths and/or overhangs. We achieve low propagation losses of $-1.36$ dB/mm and excellent mechanical stability enabling structures that approach centimeter scales, all while reducing fabrication times by $\sim90\%$ compared to full TPP fabrication of the same circuit with a constant hatching distance.

## 2 Accelerate 3D waveguide printing with flash-TPP

OPP is often employed to process thin material layers such as in standard semiconductor photo-lithography. The process is based on polymerizing a photosensitive resin using blue or UV light. The surface of the thin resin-layer is scanned or irradiated through a photomask, and the liquid resin solidifies in the light-exposed regions. Repeating this process layer-by-layer enables the fabrication of 3D structures. However, to achieve complex and truly 3D circuits, a large number of photo masks is required that also need to be precisely aligned. This has the potential to substantially reduce one of the principles motivations of 2D lithography: the efficient fabrication. Unlike OPP, photocurable resin transparent for a NIR femtosecond (fs) laser allow directly printing deep inside the resin’s volume via TPP. The basic principle of TPP relies on transforming monomer molecules located within the femtosecond writing laser’s focus into macromolecules in a solid state. For the commercially available IP family thermo-setting photoresists, i.e. IP-S, IP-Dip, IP-L and IP-G, the resin is essentially transparent from $\sim633$ nm until 2400 nm.

The Nanoscribe GmbH (Photonic Professional GT) TPP system used for printing our structures is equipped with a fs-laser operating at 780 nm, and galvanometer mirrors for rapid beam movement in the lateral directions. The femtosecond laser is tightly focused into the resin through an objective lens of high numerical aperture (25× magnification and NA = 0.8). TPP leads to the formation of a rugby-ball-shaped polymer within the focal spot, the writing voxel, and the monomer resin in its surrounding remains unmodified. The voxel-size depends mainly on the laser exposure dose, which is linked to the laser power and the galvanometer...
mirrors’ scanning speed [27]. This voxel-size is relevant when considering feature-sizes of few microns due to a diffusion process that leads to a local modification of refractive index gradients of the polymerized structures after development. In the TPP lithography process, the liquid negative-tone IP-S photoresist, with \( n \approx 1.51 \) when fully TPP polymerized [28, 29], acts as an immersion medium for the objective lens. In particular, the polymerization mechanism in the IP-S photoresist is catalyzed via a photo-initiator based on aromatic ketones, which accelerates the polymerization process by creating reactive species, i.e. free radicals, cations or anions [30]. After polymerization, the unexposed photoresist is removed in a two-step development process using propylene-glycol-methyl-ether-acetate (PGMEA) as a developer for 20 min, followed by rinsing in isopropyl alcohol (2-propanol) for 3–5 min.

One challenge of 3D TPP following the classical approach is that printing circuits with mm³ volumes quickly results in unfeasible fabrication time in excess of 20 h. However, 3D waveguide circuits are defined via the trajectories of waveguide cores, while waveguide claddings are inherently realized through the surrounding, ‘unstructured’ volume of a lower refractive index. One can therefore constrain the high-resolution yet slow TPP process to creating the waveguide cores, and use indiscriminate, hence fast, single-step blanket exposure above the resin photo-initiator’s absorption energy to develop the entire remaining chip in one flash. The working principle of flash-TPP is illustrated in Figure 1(a)–(c). Figure 1(a) depicts the usual dip-in TPP printing procedure, where the microscope objective is directly immersed into the resin. The structure printed on top of a fused silica substrate is then developed using the previously described steps. Next, we transfer the developed circuit to a UV chamber (Rolence Enterprise Inc., LQ-Box model, 405 nm wavelength, 150 mW/cm² average light intensity) as shown in Figure 1(b), and we control the OPP dosage \( D \) via the duration of the UV exposure. Here, we characterize the effects of exposure times between 0 and 60 s. The combination of DLW-TPP with UV lithography has been used by Eschenbaum et al. [31] and in Lim et al. [32] to fabricate high resolution microstructures in 3D. However, unlike flash-TPP that manipulates the refractive index of a unique photoresist, those concepts require polymerizing two different photoresists in two separate fabrication steps. Moreover, the methodology in [32] is only effective for 2D fabrication and becomes time-consuming if used for 3D fabrication due to the layer-by-layer process.

We further accelerate the fabrication process by locally and dynamically adjusting TPP writing conditions according to the functionality of each circuit’s component. Minimizing propagation losses requires minimizing the roughness at the core-cladding interface, and we use a small spacing between voxels in the \((x, y)\)-plane, i.e. with a small hatching distance \( h \). At the same time, we can maximize the vertical distance between consecutive slices, or slicing distance \( s \), which we found not to significantly affect the core-cladding interface roughness and equally accelerates the printing process. Crucially, the writing

\[ \text{Waveguide core: Hatching = 0.4 \mu m} \quad \text{Mechanical support: Hatching = 0.8 \mu m} \]

\[ \text{TPP} \quad \text{OPP} \]

\[ 50 \mu m \]

**Figure 1:** Flash-TPP for 3D integrated photonics.
(a) Illustration of the dip-in process for DLW of 3D waveguides. The IP-S resin is polymerized via TPP using a 780 nm laser and a 25X microscope objective. (b) A UV light source polymerizes the unexposed regions of the structure. The flash-TPP concept leverages both, one and two photon polymerization. (c) SEM micrograph of a 3D-printed cross-section cutting through a cuboid integrating 16 waveguides with diameters ranging from \( d \in \{0.8 : 0.4 : 6.8\} \mu m \). The waveguide cores are printed in high-resolution for maximizing optical performance, while structures only serving mechanical stability are printed with the lowest possible spatial resolution. Red colour represents the TPP-printed regions, blue colour represents regions that are exclusively polymerized with one-photon polymerization (OPP).
power needs to be adjusted in order not to overexpose the TPP-written voxels, which results in micro-explosions and high propagation losses. Other sections, such as the outer cladding of a 3D chip or internal columns for support do not interact with the optical signals, and hence they simply need to be mechanically sturdy. We therefore maximally increase the hatching distance, and since printing time scales $\propto (h^2 s)^{-1}$ this has a significant impact.

The structure in Figure 1(c) shows a scanning electron microscopy (SEM) micrograph that illustrates the overall concept. A circuit is first fabricated in a single 3D direct laser TPP lithography (red region) step, which is followed by OPP via irradiation with the UV light source (blue regions) for several seconds. Ocier et al. [33, 34] have demonstrated controllable refractive indices via beam exposure (SCRIBE), a 3D fabrication method that does not require the printing of a mechanical support since it polymerizes the photoresist inside porous thin film. However, a disadvantage fundamentally associated to this concept is that the focus quality suffers from spherical aberrations when writing large structures, as these require focusing deep inside the medium. Flash-TPP has the advantage of printing 3D microstructures using a dip-in configuration, which avoids this effect as here demonstrated by millimeter-tall yet $\mu$m-diameter waveguides. Furthermore, the SCRIBE method is limited to specific substrates such as porous silicon and silicon oxide. These types of substrates have shown inconsistencies in their optical properties for long periods (after few weeks), and especially if used in a humid environment [35]. Finally, flash-TPP has no compatibility issues with other types of surfaces (glass, metal, CMOS…) as long as adhesion of the 3D fabricated structures can be achieved.

3 Two-photon polymerization fabrication parameters

We printed a set of five free-standing waveguide cores with $20 \mu$m height and $d = 5 \mu$m diameter using a range of

![Figure 2: Structural and optical characterization of the TPP-printed waveguide cores. (a) SEM micrographs for TPP laser power LP $\in \{7, \ldots, 19\} \text{ mW} \text{ of } 5 \mu$m diameter and $20 \mu$m height waveguides, using hatching $h = 0.4 \mu$m and slicing $s = 1 \mu$m. Best combination highlighted in red. (b) Impact of hatching distance $h \in \{0.3 : 0.1 : 0.7\} \mu$m, with fixed $s = 1 \mu$m and LP = 15 mW. Best combination highlighted in red. (c) Output intensities of $d = 3.3 \mu$m diameter and $300 \mu$m high waveguides fabricated with the same hatching, slicing and LP parameters as in (b). Sample cured with UV light during 20 s. Only for the largest hatching distance one can see weaker optical confinement and scattering of the optical mode. (d) Total losses versus hatching distance $h$ for waveguides of the same diameter $d = 3.3 \mu$m and height $300 \mu$m as in (c).]
TPP laser powers (LP) and a constant hatching distance of \( h = 0.4 \, \mu m \). As globally fixed parameters in all our fabrications reported in this article we use a scanning speed of 10 mm/s and a slicing distance of \( s = 1 \, \mu m \). A SEM micrograph after development is shown in Figure 2(a), with LP \( \in \{7, \ldots, 19\} \) mW. Waveguide cores shown in the first two images were printed with LP = 7 mW and LP = 11 mW, which results in rough and inhomogeneous surfaces. Increasing the laser power to 15 mW leads to larger TPP voxels and consequently a smoother surface. In contrast, exceeding LP = 15 mW leads to overpolymerized waveguides and burning of the polymer, see the last two images of Figure 2(a). We therefore select LP = 15 mW and proceed to optimize the second parameter by scanning the hatching distance from \( h \in \{0.3 : 0.1 : 0.7\} \, \mu m \), with results shown in the SEM micrographs of Figure 2(b). The first two micrographs, with respectively \( h = 0.3 \, \mu m \) and \( h = 0.4 \, \mu m \), reveal smooth surfaces compared to the last three. We therefore select \( h = 0.4 \, \mu m \) for the following investigations because, first, the visual inspection of the surface quality shows only a negligible difference compared to \( h = 0.3 \, \mu m \), but using \( h = 0.4 \, \mu m \) reduces the fabrication time by a factor 1.8. Second, for \( h = 0.3 \, \mu m \) results were not always reproducible, a finding we attribute to spontaneous micro-burnings within waveguide cores as the smaller hatching distance increases the accumulated TPP-irradiation. SEM analysis showed that for these parameters the diameter of waveguide cores exceed the design diameter \( \tilde{d} \) by \( d_0 = 0.5 \, \mu m \) due to the non-negligible TPP voxel-size. Hence, for the following investigations we consider the effective diameter \( \tilde{d} = d + d_0 \).

Finally, to carry out an initial evaluation of the propagation losses on the hatching distance \( h \), we have characterized five printed waveguides with length of 300 \( \mu m \) at \( \lambda_0 = 660 \, nm \) wavelength with identical parameters as in Figure 2(b). Figure 2(c) shows the output intensities of the waveguides printed with \( h \in \{0.3 : 0.1 : 0.7\} \, \mu m \) at a constant diameter of \( \tilde{d} = 3.3 \, \mu m \) with LP = 15 mW. After development the samples were UV cured during 20 s. The first four outputs have practically identical intensity distribution, and a notable reduction in optical confinement can only be found for \( h = 0.7 \, \mu m \). This can also be seen in Figure 2(d), which characterizes overall optical losses (injection, propagation, outcoupling) on \( h \). Overall optical losses slowly grow with \( h \) until they dramatically increase for \( h = 0.7 \, \mu m \). We consequently select \( h = 0.4 \, \mu m \) and LP = 15 mW for the fabrication of the TPP-printed waveguide cores, mechanical support structures were printed with low-resolution (\( h = 0.8 \, \mu m \)) to reduce printing times. We introduced walls between rows of waveguides in order to maintain a flat top-surface of the cuboids to counteract the effect of shrinkage during sample development, which otherwise results in buckling of the waveguides.

### 4 Modal confinement

We 3D-printed cuboids embedding 16 waveguides with diameters ranging from \( \tilde{d} \in \{1.3 : 0.4 : 7.3\} \, \mu m \) and height of 300 \( \mu m \). We use UV exposure times for the OPP of 0, 5, 20 and 60s, which results in irradiation doses \( D \) of 0, 750, 3000 and 9000 mJ/cm\(^2\), respectively.

By fitting the experimental output intensities for different diameters, we extract the waveguide’s normalized frequency \( V = \frac{d}{\tilde{d}} \tilde{d} \, NA \), where NA = \( \sqrt{n_1^2 - n_2^2} \) is the numerical aperture and \( n_1 \approx 1.51 \) and \( n_2 \) are the refractive indices of core and cladding, respectively. Each intensity is fitted to the fundamental LP\(_{01}\) mode for diameters below the cut-off condition for a second propagating mode, as shown in Figure 3(a) for an exemplary waveguide of \( \tilde{d} = 4.5 \, \mu m \) that was UV cured with \( D = 3000 \, mJ/cm^2 \). The LP\(_{01}\) mode intensity profile in step-index (STIN) waveguides is described by \( I_0^0(u/v) \) for \( |r| < \tilde{r} \) and \( K_0^0(v/r) \) for \( |r| > \tilde{r} \), where \( \tilde{r} = \tilde{d}/2 \) and \( u, v \) the modal parameters [22]. Therefore, we individually determine the normalized frequency \( V = \sqrt{u^2 + v^2} \) for each waveguide and Figure 3(b) shows the experimental characterization for all effective waveguides diameters \( \tilde{d} \) cured using different UV doses \( D \). It is clear that the shorter we expose the circuit, the larger the normalized frequency. From the slope of the linear regressions (dashed lines) in Figure 3(b) we obtain the average NA for each UV exposure, which is shown in Figure 3(c).

As it can be seen in Figure 3(c), the NA appears to slightly decrease for larger diameters. We attribute this to the diffusion during development, which leads to a smoother refractive index transition between waveguide core and cladding and consequently converts the STIN profile to an effective gradient index (GRIN) profile [33, 36]. This phenomenon would additionally increase the NA for small diameter waveguides. Furthermore, some UV unexposed waveguides (\( D = 0 \, mJ/cm^2 \)) are absent in Figure 3(c). The reason is that the small diameter waveguides experienced displacement, either directly after fabrication or after several weeks (>2 weeks).

Finally, Figure 3(d) shows the evolution of the average (NA) and \( (n_2) \) as a function of \( D \). The excellent agreement with the fit demonstrates that we can precisely tune the numerical aperture of the different waveguides, which decreases exponentially till it reaches a plateau. Therefore, considering the refractive index of the core is approximately constant once
fully TPP-polymerized [26], all variations of NA versus exposure dose $D$ can be assigned to the refractive index of the cladding following $n_2 = \sqrt{n_1^2 - NA^2} = A - B \exp(\kappa D)$, by which we obtain $\kappa = 6.7 \times 10^{-4} \text{ mJ}^{-1} \text{ cm}^2$.

5 Propagation losses

To evaluate losses we fabricated a set of waveguides with lengths spanning from 0.1 to 6 mm. The diameter is $\tilde{d} = 3.7 \mu m$ and the UV expose dose $D = 3000 \text{ mJ/cm}^2$, which provide high NA while remaining single-mode. Minimizing injection losses requires adiabatically modifying the waveguide core diameter to transition the mode size of the free-space input to the one of the waveguide. The angle of such tapers, i.e. the taper-rate, needs to be sufficiently small to not excite higher order modes and hence to satisfy the adiabatic transition condition. We minimized the optical losses of a set of tapers with lengths $\epsilon \in \{10 : 10 : 50\} \mu m$ and waveguide core diameters $\tilde{d} \in \{3.7 : 0.4 : 5.7\} \mu m$ in order to determine the best taper-rate for efficient mode coupling. These we found to be tapering from an input diameter of 4.9 $\mu m$ to the target waveguide diameter of 3.7 $\mu m$ during a taper-length of 40 $\mu m$.

Figure 4(a) depicts the total losses for the fundamental LP01 mode on a semi-logarithmic scale, and via the linear fit we determine $-1.36 \text{ dB/mm}$ propagation and $-0.26 \text{ dB}$ injection losses. Our measurements are highly reproducible across the wide range of propagation lengths we here examined, in particular considering that each waveguide longer than 0.5 mm was fabricated on a different substrate. This demonstrates the excellent reproducibility and high quality of our $(3 + 1)D$ flash-TPP fabrication as well as of our optical characterization.

Propagation losses do not yet reach the bulk absorption $\approx 0.055 \text{ dB/mm}$ of the IP-S photoresist at 660 nm [26],...
and for now are around one order of magnitude above standard silicon photonic waveguides \[37\]. Compared to single-pass DLW of waveguides in fused silica glass, our propagation losses are comparable at our characterization wavelength, while we achieve more than an order of magnitude lower injection losses \[38\]. Still, our results represent a factor 5 improvement compared to previously 3D-printed waveguides \[17\]. Figure 4(b) shows the 6 mm long waveguide printed for this study, imaged next to a match for scale.

6 Temporal stability

As a final study, we investigated aging of the 3D-printed waveguides under ‘rest’ (ambient laboratory light levels) and operation conditions (guiding optical signals), as temporal stability naturally is of vital importance for reliability. Aging could originate from such effects as thermal diffusion \[39\], shrinkage \[40\] or undesired photo-polymerization of areas with low degree of polymerization \[26\].

The flash-TPP concept relies on incomplete polymerization of the areas corresponding to waveguide cladding, hence some photo-initiators could remain active after development. If unintended post-development optical exposure is sufficient to initiate the photo-initiator’s reactions, the waveguide cladding could polymerize towards a higher degree, i.e. increasing its refractive index. Consequently, \(\Delta n\) and the related optical properties would be altered in time. To explore the temporal stability we measured the waveguide’s NA over time. All structures have been kept under standard irradiation intensity of our lab and have not been shielded from room light. As depicted in Figure 5(a) and for such ‘resting’ conditions, the NA of all waveguides explored in the previous sections do not suffer any relevant change during their continuous characterization spanning \(\sim 3000\) h.

We evaluated the degradation of our waveguides during continuous operating conditions. We examined the effect of injecting light with a power of 0.25 mW during \(\sim 600\) h at \(\lambda_0 = 660\) nm into a waveguide of \(d = 2.9\) \(\mu\)m, 300 \(\mu\)m height and UV exposed with \(D = 3000\) mJ/cm\(^2\). We found the optical properties to be stable within our measurement resolution, see Figure 5(b) showing the NA monitored during \(\sim 600\) h under continuous operation, with average \(\langle NA\rangle = 0.1014\) (dashed line) with a standard deviation of \(3 \times 10^{-4}\). Consequently, we confirm the reliability of the flash-TPP technique in a practical context.

Finally, we also evaluated the waveguide’s resilience to direct sun exposure, for which we placed one sample on a window sill during summer months in central Europe. This resulted in the almost complete polymerization of the cladding area after \(\sim 15\) days and hence the erasure of the light-guiding structures. The direct and unprotected UV pollution

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4}
\caption{Optical losses and large scale integration.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5}
\caption{Temporal stability.}
\end{figure}
radiation from the sun therefore appears to be sufficiently strong [41] to launch the photo-initiators reaction and thus the polymerization process. We also examined the effect of direct sun exposure on waveguides for which the cladding was printed with a low TPP power dose (i.e., 1, 2 and 3 mW), and no difference compared to the flash-TPP was found; the structures equally got erased by the direct sun exposure. Therefore, we can conclude that the aging under direct exposure to sun light is through the general augmentation in polymerization-degree and does not depend on the polymerization technique used during fabrication.

7 Flash-TPP printing time

Printing time is generally proportional to a circuit’s volume, and the relative time that can be saved due to flash-TPP depends on the ratio between the areas requiring TPP and OPP exposure. Here, we assume that this ratio remains constant along a circuit’s z-positions. In previous work on (3+1)D printing we found that waveguides with a radius of \( r \approx 2.5 \mu m \) need to be separated by \( l \approx 6 \mu m \) of cladding in order to have negligible interaction [9]. Cross-talk essentially depends on the relationship of confinement and separation, and scaling of \( l \) and \( r \) should therefore remain comparable for integrated single-mode circuits with a similar \( \Delta n \). Using OPP to print the entire structure requires a fabrication time \( T_{\text{TPP}} \propto (l + 2r)^{3/2} \), while flash-TPP requires \( T_{\text{flash}} \propto \pi r^2 \). As a result, the relative duration \( \Gamma \) of flash-TPP relative to classic TPP is given by

\[
\Gamma = \frac{\pi r^2}{(l + 2r)^{3/2}} = \frac{1}{4} \left( \frac{\pi}{1 + 1/2r} \right)^{3/2},
\]

which is also the filling factor of waveguide cores within a 3D integrated photonic chip. Therefore, even for a circuit volume with maximal density of waveguide cores, the usual objective of circuit integration, using \( l \) and \( r \) defined above the fabrication time is reduced to \( \Gamma \approx 16\% \) compared to using TPP alone. This agrees with our experience, where using flash-TPP reduces the printing time to only \( \Gamma \approx 10\% \) compared to classical TPP. As a clear example, printing our 6 mm macroscopic structure exclusively only using TPP takes \( \sim 24 \) h, whereas this decreased to \( \sim 3 \) h (\( \Gamma \approx 12\% \)) using flash-TPP.

Tall waveguides require the printing of an outer cladding that serves as a mechanical support. Generally, printing mechanical support will be the limiting factor for a small number of waveguides. However, this becomes less relevant for large number of waveguides. Ultimately, the ratio between printing waveguides and mechanical support can be described using similar considerations as the simple speed factor in Eq. (1). For waveguides with radius \( r \) and separation of \( l \), the area for a waveguide core is \( A_{\text{core}} = \pi r^2 \). Using thickness \( t \) for the mechanical support, and a circuit with \( N_c \) (\( N_t \)) columns (rows), plus a mechanical support wall every \( k \) rows with thickness \( at \), the area of a circuit’s mechanical support is:

\[
A_{\text{mech}} = t \left[ 1 + \frac{1}{l} \left( N_c + N_t + a \frac{N_c N_t}{k} \right) \right].
\]

To obtain the ratio \( R \) of printing time between core and mechanical support we normalize by \( A_{\text{core}} \) and assume a square circuit profile \((N_c = N_t = \sqrt{N})\) and hatching distances \( h_m \) and \( h_c \) for mechanical support and core, respectively, we obtain:

\[
R \propto \frac{h_c}{h_m} \frac{t^2}{\pi r^2} \frac{1}{\sqrt{N}} \frac{2}{\sqrt{N}} \frac{a}{k}.
\]

For large circuits, the additional time for fabricating the mechanical support therefore reduces to

\[
R|_{N \to \infty} \propto \frac{h_c}{h_m} \frac{t^2}{\pi r^2} \frac{a}{k}.
\]

This ratio is constant and depends on the relative ratio of core and mechanical hatching distances \( \frac{h_m}{h_c} \), of the ratio between the areas of the mechanical support’s unit cell and a waveguide core \( \frac{A_{\text{mech}}}{A_{\text{core}}} \), and of the properties of the intra-wall support \( \frac{a}{k} \). In our case this is \( R|_{N \to \infty} \propto \frac{0.8 \mu m^2}{\frac{0.4}{7.1 \mu m^2}} = 3.52 \), and printing mechanical support and waveguides therefore takes comparable amounts of time. However, we spaced individual waveguides far apart (30 \( \mu m \)). Using the dense fabrication with \( l = 6 \mu m \), one can most likely remove the mechanical support, and in this case the scaling is with \( R|_{N \to \infty} \propto \frac{1}{\sqrt{N}} \), hence the mechanical support takes less and less time relative to the photonic guiding structures.

8 Discussion and conclusion

We have developed flash-TPP, a simple and fast (3+1)D lithography configuration. This novel manufacturing methodology is based on combining DLW-TPP and OPP, here demonstrated with the commercially available IP-S photoresist, for the fabrication of polymer-cladded single-mode 3D optical waveguides. The flash-TPP concept synergistically combines three principles. First, waveguide cores are printed via DLW-TPP using a precisely calibrated TPP laser power and with high horizontal resolution by minimizing the hatching distances between voxels \((h = 0.4 \mu m)\), ensuring low propagation losses due to smooth interfaces. Second, areas purely serving as mechanical support do
not interact with the guided wave and no special control of the spacing between TPP-voxels is needed. These are consequently printed with a large hatching distance \( h = 0.8 \mu m \). Finally, instead of building the entire cladding by DLW-TPP, we polymerize in a single instance the integrated photonic circuit via OPP using UV blanket irradiation. Combining these three aspects we are able to reduce the printing time by \( \sim 90\% \) compared to classical TPP only fabrication. Furthermore, we show that for integrated photonic chips, where the packing density is adjusted to obtain small cross-talk, one can generally expect an acceleration of fabrication on this order.

Multiple conventional photolithographic techniques are also employed to fabricate 3D structures. Projection micro-stereolithography (\( \mu \)SL) leveraging micro-continuous liquid interface production (\( \mu \)CLIP) is an excellent technique to print macroscale optical components (e.g. a 3 mm diameter aspherical lens) with high speed \( (4.85 \text{ mm}^3 \text{ h}^{-1}) \) [42]. Flash-TPP achieves \( 0.014 \text{ mm}^3 \text{ h}^{-1} \), which is excellent compared to the \( 0.002 \text{ mm}^3 \text{ h}^{-1} \) of TPP alone. Importantly, \( \mu \)SL-\( \mu \)CLIP does not allow the high-resolution refractive index control required for single-mode waveguides or high-resolution volume holograms. Moreover, the highest feature resolution obtained with \( \mu \)SL-\( \mu \)CLIP is \( \sim 50 \mu m \) [43], whereas for flash-TPP we readily reach \( \sim 1.3 \mu m \) with this particular resin. High-quality surface (\(< 7 \text{ nm}\)) using \( \mu \)SL [44] requires a sequence of steps, while we achieve smooth surface (\( \sim 20 \text{ nm roughness} \)) in a single fabrication step. Flash-TPP is therefore an excellent candidate for manufacturing large scale complex and high-resolution GRIN elements with excellent quality. Finally, flash-TPP could be further accelerated by multiplexing the single into multiple writing voxels using phase modulation with a spatial light modulator [45]. This technique can directly be combined with our flash-TPP method, hence both concepts are highly complementary.

We achieved precise control over the refractive index difference \( \Delta n \) between waveguide core and cladding by adjusting the UV exposure dose during our blanket OPP illumination, here ranging from 0 to 9000 mJ/cm². We fitted the waveguide’s output intensities to the LP\(_{01}\) mode of waveguides for variety of UV exposure doses and diameters below the cut-off condition of the second LP mode. From these fits we obtain a robust characterization of a waveguide’s NA as a function of the UV dosage, and our data shows an excellent agreement with the exponential trend expected for saturation processes such as UV curing, obtaining numerical aperture values ranging between NA = 0.10 and NA = 0.16. We finally determined low propagation losses of \( \sim 1.36 \text{ dB/mm} \) and a continuous characterization of our 3D-printed waveguides spanning \( \sim 3000 \text{ h} \) demonstrated their temporal stability. Importantly, we demonstrated the reliability of flash-TPP under operating conditions simulating by continuously injecting 0.25 mW. During \( \sim 600 \text{ h} \) we did not find any deterioration. In DLW of waveguides in fused silica, post fabrication thermal annealing reduced losses from similar levels down to \( \sim 0.08 \text{ dB/cm} \), which could be a promising avenue for approaching the material’s absorption limit.

For 3D printing complex shapes, UV exposure might not be homogeneous due to ‘shadows’ cast by TPP structures. Should this prove to be a challenge, one could consider UV exposure from different sources placed at various locations around the sample. Furthermore, the UV exposed sample can be post-cured in order to accelerate and homogenize the polymerization process [26]. Strong absorption at 405 nm of the liquid IP-S resist can attenuate OPP at mm-depth inside a large-scale integrated circuit. We did not observe this effect in our samples, most likely due to their high-aspect which enables UV-exposure through the nearby side-walls. Uniform OPP exposure levels in the volume of mm-scale circuits of aspect ratio near unity could be achieved by shifting the OPP wavelength closer to the edge of absorption of the photo-initiator.

Overall, we have established (3 + 1)D flash-TPP as a powerful manufacturing technique towards the fabrication of polymer-based 3D integrated photonic circuits. The concept can be particularly advantageous in applications requiring integrated photonics circuits of complex 3D topology that render a standard 2D integration impossible. Two-photon grayscale lithography now targets mass fabrication of 2.5D microstructures using the new generation of 3D printers [46], which pushes scanning speeds from the previous 0.625 m/s to 6.25 m/s. This new generation shows the excellent scaling potential of the fabrication technique, and flash-TPP can immediately be transferred to this concept.

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