# Laser-based microscale patterning of biodegradable polymers for biomedical applications

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**Abstract:** In this paper, we throw light on current state of advancements in the field of biodegradable polymers and devices targeted at biomedical applications. A short introduction involving biodegradable polymers, various fabrication techniques for patterning biodegradable polymers and an extensive coverage of ultraviolet (UV) laser micromachining of polymers is presented. We also present UV laser micromachining of biodegradable polymers for potential biomedical applications. Micro-holes and microchannels, which are important features in many micro-devices, have been etched on biodegradable polymers by UV lasers. The morphology and surface conditions of the etched micro-featuress in the polymers are studied using SEM and surface profilometre. The effect of laser parameters such as laser wavelength and pulse energy on the etching quality is also investigated. This work on using UV laser for ablation of biodegradable polymers is the first of its kind.

**Keywords:** biodegradable polymers, biomedical applications, bioMEMS, laser ablation , laser micromachining, photochemical, photothermal, UV lasers .

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**Biographical notes:** Shaochen Chen received his PhD degree in mechanical engineering from the University of California at Berkeley in 1999. His research areas include laser-based micro- and nano-manufacturing, micro-electro-mechanical systems, and thermal/fluid transport in micro- and nano-systems. His research applications include computer and microelectronics manufacturing, biomedical engineering, and life science. Vijay Kancharla and Yi Lu are graduate students with research areas in laser material processing using UV and ultra fast lasers. Their current research involves laser micromachining of biodegradable polymers by UV lasers for potential applications in fabricating biodegradable micro-devices.

## 1 Introduction

Implantable medical devices are currently fabricated from non-biodegradable materials like silicon, glass, and plastics [1, 2]. One of the major problems associated with these non-biodegradable devices is that they must be removed surgically once the drug is exhausted. Biodegradable polymers hold great promise as new materials for implantable biomedical micro-devices, due to their biocompatibility and natural

ability to degrade in tissue over time. The advances in biodegradable polymer science are paving way for fabrication of implantable biodegradable micro-devices such as drug delivery systems, tissue engineering, and catheters in the field of biomedical engineering [3, 4]. Some of the commercial biodegradable devices available are sutures, dental devices, orthopedic fixation, and guided tissue.

### *1.1 Biodegradable polymers*

Biodegradable polymers can be either natural or synthetic. In general, synthetic polymers offer greater advantages than natural materials in that they can be tailored to give a wider range of properties [5]. The general criteria for selecting a polymer for use as a degradable biomaterial are to match the mechanical properties and the time of degradation to the needs of the application. The ideal polymer for a particular application would be configured so that it:

1 Has mechanical properties that match the application, remaining sufficiently strong until the surrounding tissue has healed.

2 Does not invoke an inflammatory or toxic response.

3 Is metabolized in the body after fulfilling its purpose, leaving no trace.

4 Is easily processable into the final product form.

5 Demonstrates acceptable shelf life.

6 Is easily sterilized.

The factors affecting the mechanical performance of biodegradable polymers include monomer selection, initiator selection, process conditions, and the presence of additives [6]. These factors in turn influence the polymer's hydrophilicity, crystallinity, melt and glass-transition temperatures, molecular weight, molecular-weight distribution, end groups, sequence distribution, and presence of residual monomer or additives. In addition, the biodegradable materials must be evaluated for each of these variables for its effect on biodegradation [7]. A list of typical biodegradable polymers in biomedical engineering is shown in Table 1.

The research work integrating biopolymers, lasers and other fabrication techniques have huge potential applications in the biomedical field. Johnson *et al.* [8] reported on fabrication of microchannels using both hot imprinting and UV laser

**Table 1** List of some biodegradable polymers for biomedical applications.

| Polymer | Melting point (°C) | Glass-transition Temp (°C) | Modulus (Gpa) | Degradation time (months) |
|---|---|---|---|---|
| PGA | 225∼230 | 35∼40 | 7 | 6–12 |
| LPLA | 173∼178 | 60∼65 | 2.7 | >24 |
| DLPLA | Amorphous | 55∼60 | 1.9 | 12–16 |
| PCL | 58∼63 | (−65)∼(−60) | 0.4 | >24 |
| PDO | N/A | (−10)∼0 | 1.5 | 6–12 |
| PGA-TMC | N/A | N/A | 2.4 | 6–12 |
| 85/15 DLPLG | Amorphous | 50∼55 | 2.0 | 5–6 |

ablation on PMMA polymer for potential applications to realize lab-on-a-chip devices. Initial studies on laser etching of biodegradable polymers by Kancharla and Chen [9] revealed that UV laser is a promising tool for micro-patterning biodegradable polymers for bioengineering applications. Armani *et al*. [10] proposed three unique micro-patterning techniques for fabricating bio-implantable drug delivery chip using PCL polymer. Porous biodegradable polymers are extensively used as tissue analogue scaffolds in cellular transplantation therapies for organ failure, and a novel method was introduced by quantitating the void and surface microstructure of porous biocompatible polymers using confocal laser-scanning microscopy [11]. A diode laser of 806 nm was employed by Sorg and Welch [12] for tissue soldering using biodegradable polymers. Toenshoff *et al*. [13] presented femtosecond laser micromachining of biodegradable polymers for applications in cardiovascular implant devices.

### *1.2 Overview of micro-patterning techniques*

Micro-patterning of polymeric materials is conducted by many methods and techniques. However, techniques such as electron beam etching produces too much heat and damage the polymers while photolithography involves multiple process steps and chemical solvents in the process [14]. Properties of biodegradable polymers degrade easily with chemical solvents or thermal impact. Moreover after fabrication the residual chemical agents on the polymer surface could be toxic to the cells if using chemical etching. Atomic force microscopy (AFM) utilizes mechanical forces to etch the surface. However, this method is very slow and cannot be used conveniently to etch large surface area [15]. Non-photolithographic techniques, such as soft lithography techniques, have been developed for polymer microfabrication [16]. Soft lithography techniques include microcontact printing, micromoulding in capillaries, microtransfer moulding and replica moulding, imprinting, and injection moulding. However, the utility of these techniques is often limited by the availability of appropriate masters. Some techniques of polymer ablation have been summarized in Table 2, considering the various factors that effect the polymer ablation.

**Table 2** Summary of micromachining processes for polymers.

| Processes | Thermal effects (HAZ) | Material processing speed | Process steps | Cost | Chemicals involved | Mechanism involved | Scan area |
|---|---|---|---|---|---|---|---|
| Photolithography | Less | Fast | Multiple | High | Yes | UV light and masks | Large |
| Electron beam etching | High | Slow | Single | High | No | Beam of electrons | Small |
| Atomic force microscopy | Less | Very slow | Single | High | No | Mechanical forces | Very small |
| Soft lithography | Less | Fast | Multiple | Low | No | Mould transfer | Large |
| Laser micromachining | Medium | Fast | Single | Medium | No | Photon energy | Large |

### 1.3 Lasers for micro-patterning

Laser micromachining makes it possible to pattern biodegradable polymers on the microscale, unlike the difficulties associated with the above techniques. Photons of the laser light act as 'clean particles,' and the laser irradiation is essentially non-invasive and a single-step process. The most popular lasers used for machining polymers are the UV lasers. The most popular and commercial UV lasers include excimer, argon-ion, tripled and quadrupled Nd: YAG, fluorine, helium-cadmium, metal vapour, and nitrogen [17]. Among these, excimer lasers (193 nm to 351 nm) are the best choices, but Nd: YAG lasers are considered if they are Q-switched and frequency-tripled to produce 266 nm wavelength. Excimer lasers are extensively used for photoablation, chemical etching, lithography, and surface cleaning. The benefits of excimer lasers are attributed to their short wavelengths (193 nm to 351 nm), high energy per pulse, and nanosecond (ns) pulse widths. The small wavelengths allow strong interactions of the beam with a variety of work piece materials. Laser micromachining with nanosecond green or infrared lasers has strong thermal effects [18], whereas UV lasers can remove material through direct solid-vapour ablation. The incident photon energy is high enough to break the chemical bonds of the target material directly, the material is dissociated into its chemical components, and no liquid phase transition occurs in this process. This photochemical process has much minimized heat effects compared with the photothermal process. This important feature makes UV laser micromachining very attractive for biodegradable polymer materials, since thermal damage to the non-machined part can be minimized.

### 1.4 Laser ablation of polymers

Laser ablation of polymer surface is a function of the energy deposited in the solid in unit time. It was reported that when UV pulses strike the surface of the polymer, depending upon the wavelength and laser intensity, 0.01–0.1 micron of the material is etched away [19]. The ablation is accompanied by ejection of matter in the form of species such as atoms, molecules, ions, and clusters because of the interaction of an intense laser pulse with the polymer material. The macroscopic effects of ablation include plasma, acoustic shocks, and cratering of the surface [20]. The capability of UV laser to ablate a polymer depends on the absorption characteristics of the polymer at that wavelength. Some of the general features of UV ablation of polymers are summarized below [19–28]:

1 Polymer ablation takes place within 10–100 nanoseconds for a nanosecond laser beam.

2 For fluences near or above the ablation threshold, the etch depth follows Beer-Lambert's law (photochemical, linear absorption). For fluences far above the threshold, thermal effects contribute to the etch depth. In addition, the longer the wavelength is, the stronger are the thermal effects.

3 Wavelength affects absorption and threshold fluence. The etch depth per pulse is smaller for a weaker absorber (lower absorption coefficient) than for a stronger absorber.

4 The formation and expansion of the plasma plume during the laser pulse characterize the rapid etching process. The etch depth per pulse increases with

energy fluence until the phenomenon of saturation is reached. 'Saturation' is a mechanism involving the blocking of the trailing part of the laser pulse by both the plume and the excited polymer species generated by the leading part of the pulse. This occurs only at high energy densities and prevents additional material removal.

5 Ablation is accompanied by an acoustic signal that decreases with increasing laser wavelength.

6 The relaxation rate, the period of time in which the excited states of the polymer endures, affects the absorption of the laser light by the material. If the relaxation rate is too slow compared to the excitation rate, the bleaching or blocking effect occurs and reduces the absorption of laser energy.

7 Ablation takes place in the temperature ranging from 400 to 800 °C.

8 The small absorption depth coupled with short laser pulses and low thermal conductivity of polymers restricts the extent of heat transfer, leading to precise material removal and a small heat-affected zone.

The absorption of photons in the polymer occurs to a depth determined by the absorption coefficient ($\alpha$) of the material. This absorption coefficient is obtained by Beer–Lambert's law [21], $I = I_o \exp(-\alpha x)$, where $I$ is the laser intensity, $I_o$ is the laser peak intensity, and $x$ is the penetration depth. The threshold fluence is the intensity at which the ablation starts, below this intensity the energy of photons is absorbed as heat in the polymer surface. For fluences far above the threshold, thermal effects contribute to the etch depth. In addition, the longer the wavelength, the stronger are the thermal effects. The two fundamental mechanisms involved in the laser ablation are believed to be photochemical and photothermal mechanism. In photochemical mechanism the photon energy of the light is used to break the chemical bonds of the polymer directly whereas in photothermal mechanism the material is ablated by melting and vapourizing [22, 23]. For photochemical ablation to occur, energy of the photons at that wavelength should overcome the intermolecular bond energies of the polymer (Table 3). The relation between the photon energy of light and laser wavelength is given by $E = 1.245 / \lambda$, where $\lambda$ is the laser wavelength and $E$ is the photon energy. The photon energy decreases as the wavelength increases. It can be seen that for photochemical ablation to occur in polymers the photon energy of the light should be greater than the bond energy of the material.

Several photochemical and photothermal ablation models have been proposed by various researchers for laser assisted polymer ablation. However, there is no concrete model for the exact ablation mechanisms involved. Some of the models presented are reviewed below. A photochemical model based on a volume change of the material after the photolysis, induced by the UV radiation is presented by Srinivasan *et al.* [19]. A thermal model was presented that a very high temperature

**Table 3** Chemical bond energies for polymers.

| Polymer bonds | H-H | O-O | N-N | C-C | C=C | C≡C | C-H | C-N |
|---|---|---|---|---|---|---|---|---|
| Bond energy (eV) | 4.48 | 5.12 | 9.76 | 3.62 | 6.40 | 8.44 | 4.30 | 3.04 |

was predicted for small etch depth per pulse ablation rates [23]. A combined photochemical-thermal model was developed in which a thermal contribution to etching is added to the photochemical contribution derived from low fluence measurements [24]. Several other theories to explain the ablation mechanism have surfaced considering issues such as absorption of light in three state chromophores [25]. Furzikov proposed a theory of the ablation mechanisms considering issues such as refractive index of polymer, microroughness, random polarization, and weak focussing of the beam [26]. Other theoretical models include dynamic model, receding surface treatment, chromophore bleaching, plume screening, and Arrhenius-type thermal activation [27, 28].

## 2 Experimental procedure and materials

Polyvinyl alcohol (PVA), poly-D-lactic acid (PDLA) and poly (ε-caprolactone) (PCL) are the biodegradable polymers used in this study. Polystyrene (PS) is also used in this work for comparison. PVA is a water-soluble polymer and has excellent physical properties. It is used in a wide range of applications such as adhesives, fibres, textile, paper sizing, and water-soluble packaging. PDLA is a biodegradable polymer with extensive medical applications due to its biodegradable property and is proven harmless to human body cells. PDLA has been used as a substrate material for potential applications in nerve regeneration in the field of tissue engineering [29]. PCL, an aliphatic polyester is one of the most important biodegradable polymers in medicine. Some of the applications of PCL are sutures and biocompatible medical devices. These polymers have unique features such as controllability of mechanical properties, tailoring of degradation rates, and minimal toxicity and immune response that make them ideal for medical uses. Micro-patterning these biodegradable polymers as substrate materials could provide guidance to the neurons at the cellular level surfaces [29].

The micromachining set-up for biodegradable polymer ablation consists of four main parts: a laser system, a beam delivery system, a micrometre-resolution *x*-*y* sample stage, and an on-line monitoring system (Figure 1). The beam delivery system consists of a mask, field lens, turning mirrors and an imaging lens to the beam for micromachining. The energy of the beam was measured at the laser output.

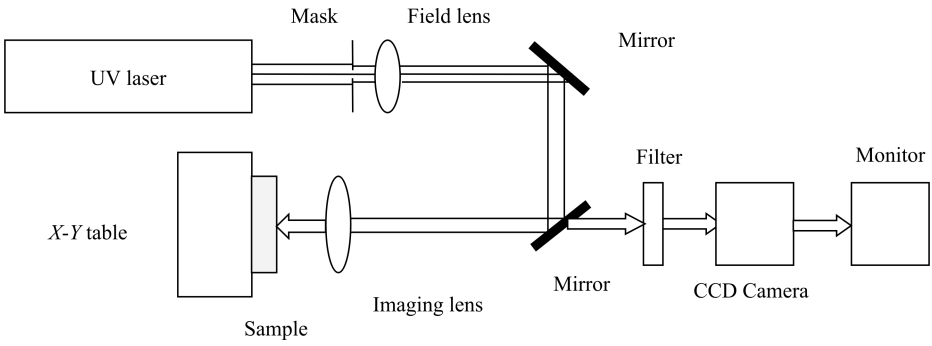



**Figure 1** Schematic setup of the laser micromachining system.

**Table 4** Properties of lasers used in this study.

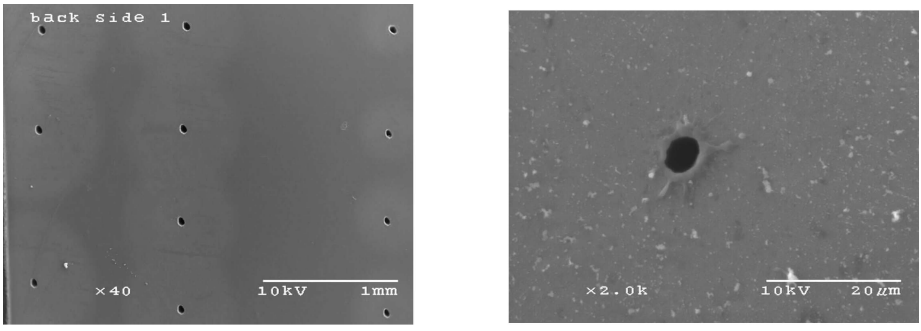
| Medium | ArF | Nd: YAG | XeCl |
|---|---|---|---|
| Wavelength (nm) | 193 | 266 | 308 |
| Pulse Energy (mJ) | 5 | 275 | 300 |
| Repetition rate (Hz) | 10∼100 | 1∼10 | 1∼100 |
| Pulse width (ns) | 6 | 6∼7 | 20 |

A spherical imaging lens was used to drill micro-holes, and a cylindrical lens was used to produce micron-size channel type patterns. A charged-coupled device (CCD) camera coupled with a TV monitor is used to provide online machining information.

A XeCl excimer laser (Lambda Physik 308 nm) and an ArF excimer laser (193 nm) were used to irradiate the polymers to fabricate microchannels. The initial beam size of $10 \times 20\,mm^2$ was chopped down using a rectangular mask to shape the excimer laser beam to obtain straight and uniform channels. Lenses with 1-inch and 2-inch focal lengths were used to obtain microchannels with different width grooves. The samples were ablated at the focal point as well as at the defocused area. A frequency quadrupled Nd: YAG at 266 nm was used to fabricate micro-hole array. A circular mask was used to shape the beam. All the experiments were conducted in ambient air and at 1 Hz pulse repetition rate. Details of the lasers used in this work are listed in Table 4.

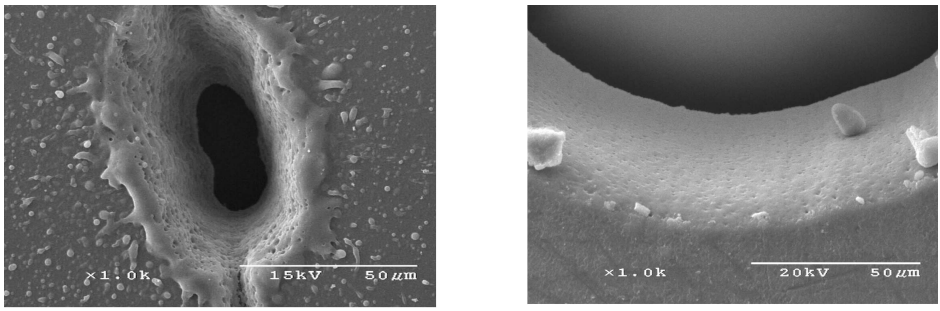
## 3 Results and discussion

Micro-holes were percussion drilled on PVA polymer using an Nd:YAG laser for its Gaussian beam profile. Arrays of holes (Figure 2a) were drilled for potential applications in biomedical engineering, such as degradable microfilters. The holes at the exit were around 5 microns in diametre and holes observed at the entry were larger. Figure 2b shows the 5 micron hole at the exit in the PVA polymer. At 266 nm wavelength, polymer melting and resolidification was noticed around the machined




(a) (b)

**Figure 2** (a) Nd: YAG laser drilling of micro-holes on PVA (b) 5 micron hole at the exit of PVA polymer.




(a) (b)

**Figure 3** Micro-hole quality at (a) 266 nm and (b) 193 nm etched in PVA.

holes (Figure 3a). For biomedical applications this is not desirable. To minimize these thermal effects a 193 nm wavelength laser was used to drill holes. From the SEM picture in Figure 3(b), no melting or redeposition was noticed. The irregularities in the hole shape can be attributed to the spherical aberration and non-circularity of the original laser beam. Using advanced beam delivery systems and beam shapers, the irregularity of the hole shape can be corrected.

Microchannels were etched on PVA, PDLA, PCL, and PS polymers. Excimer lasers (KrF and XeCl) have been used for this study. It is observed that, below the threshold energy there is no sign of ablation and above the threshold the ablation depth increases as the laser energy increases, following the Beer-Lambert's law for the laser energy used in this study [31]. At 308 nm wavelength, the absorption coefficient was found out to be $0.4 \times 10^{-4}\,\mathrm{cm}^{-1}$ and 266 nm it was found out to be $0.845 \times 10^{-4}\,\mathrm{cm}^{-1}$. An array of well-patterned microchannels on PVA can be seen in Figure 4.

To study the effect of photochemical and photothermal mechanisms on polymers at different wavelengths, the polymers have been etched at both 193 nm and 308 nm wavelengths. Channels on the polymers have been etched using the same experimental parameters and then examined under a SEM for material analysis.

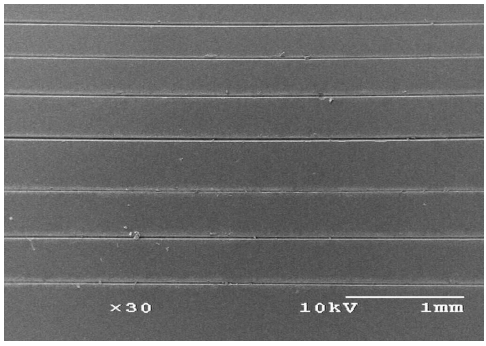
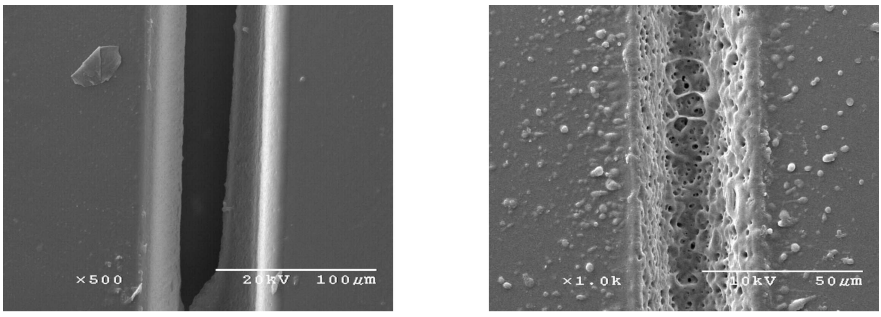
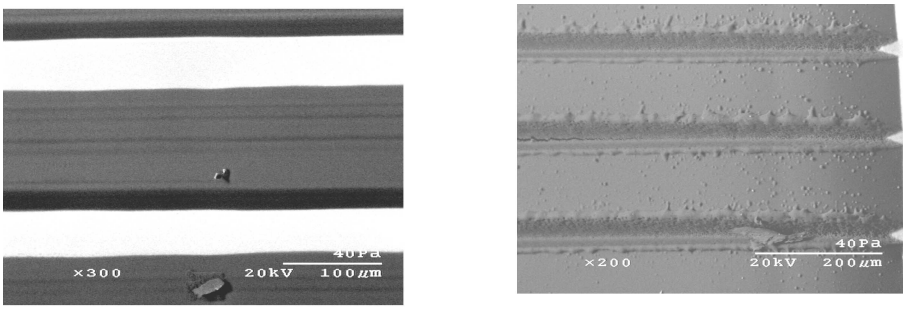



**Figure 4** Array of microchannels etched in PVA.



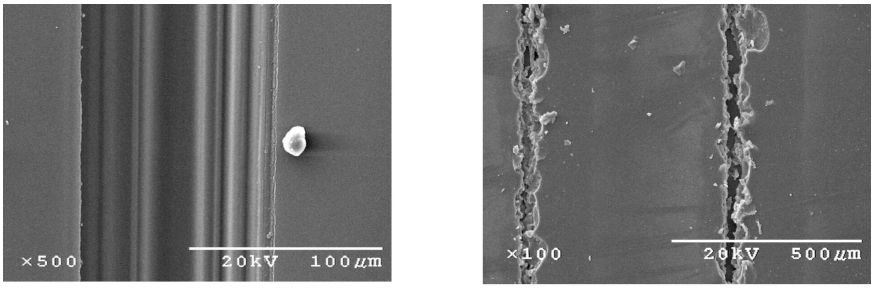

(a) (b)

**Figure 5** Microchannels in (a) PVA at 193 nm, (b) PVA at 308 nm wavelengths.




(a) (b)

**Figure 6** Microchannels in (a) PDLA at 193 nm, (b) PDLA at 308 nm wavelengths.




(a) (b)

**Figure 7** Microchannels in PS (a) At 193 nm, (b) At 308 wavelengths.

Figures 5, 6, 7, 8 show the effect of ablation mechanisms at 308 and 193 nm wavelengths for polymers PVA, PDLA, PS and PCL. It is observed that at 308 nm wavelength the effect of photothermal mechanism is very evident. This is because at 308 nm wavelength, the photon energy of light is only 4.02 eV. This energy is not

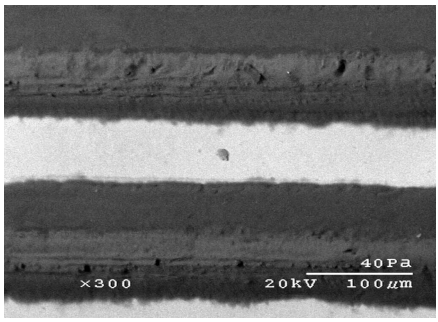



**Figure 8** Microchannels in PCL at 193 nm wavelength.

sufficiently high to break the polymer bonds, so thermal contribution for ablation plays a role in ablating the material. At 193 nm wavelength, the photon energy of the laser beam reaches about 7.9 eV, which is well above the bond energies of the polymer. Therefore the photochemical effect dominates the laser ablation process, resulting in clean cut along the wall.

## 4 Conclusions

A review of current state of advancements in the field of biodegradable polymers and devices is presented. The authors have employed laser micromachining techniques for etching biodegradable polymers for potential applications in biomedical engineering. Micron-sized channels and holes are etched successfully in biodegradable polymers, which would be an integral part of many biodegradable micro-devices. The effect of laser parameters such as wavelength, pulse energy, pulse duration on the etching quality were studied. The laser wavelength plays an important role in material ablation. For biodegradable polymers used in this work, the longer the laser wavelength, the stronger are the photothermal effect. It is concluded that deep UV lasers such as 193 nm wavelength are better choices in order to minimize the photothermal effect, resulting in direct bonding breaking and clean laser cutting.

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