Fabrication of Metallic Heat Exchangers Using Sacrificial Polymer Mandrils

Francisco Arias, Scott R. J. Oliver, Bing Xu, R. Erik Holmlin, and George M. Whitesides, Member, IEEE

Abstract—This paper demonstrates the use of poly(dimethylsiloxane) (PDMS), polyurethane (PU), epoxy, and poly(methyl methacrylate) (PMMA) as mandrils to fabricate metallic heat exchangers having 300–700 μm internal channels. The mandrils were prepared using two soft lithographic techniques—replica molding, and microembossing. To fabricate the heat exchangers, the polymeric mandrils were coated with a thin layer of metal by thermal evaporation or sputtering; this layer acted as the cathode for electrodeposition of a shell of nickel or copper that was 100 μm thick. The polymers were removed by burning them out at 400 °C in air, or by dissolving them with a tetrahydrofuran solution of tetrabutylammonium fluoride. Studies of heat dissipation showed that the nickel heat exchangers with features that range in size from 150–750 μm have thermal resistances ranging from 0.07 to 0.12 °C W⁻¹ cm at flow rates of water of ~20 L h⁻¹ and pressures of 8.6–83×10⁻³ N m⁻². [541]

Index Terms—Electrodeposition, fabrication, heat exchangers, microelectromechanical devices, molding, soft lithography.

I. INTRODUCTION

This paper describes the use of soft lithography[1], [2] in the fabrication of metallic heat exchangers containing micrometer-scale channels. Small, high-performance heat exchangers are required to cool a number of different types of devices, including microprocessors, lasers, and devices exposed to intense photon fluxes. The process described here was designed for simplicity; it was also designed to avoid processes – silicon etching, titanium diffusion bonding, tin/lead soldering, and micromachining of copper – commonly used in fabricating such devices. [3]–[5] These heat exchangers can withstand high coolant pressures (≤2.1×10⁵ N m⁻²) without fracturing because they are prepared in one piece; joining methods such as soldering or welding, which may yield structural regions with low strength, are not required.

The general fabrication process involves six steps: 1) Printing a design of these heat exchangers on a transparency. 2) Using the transparency as the photomask for photolithography to make the master. 3) Making polymer mandrils by replica molding or microembossing; 4) Depositing a thin layer of metal on the polymer surfaces by evaporation or sputtering. 5) Electrodepositing nickel on the polymeric mandril to give metallic microstructures. 6) Dissolving or incinerating the polymeric mandrils to yield metallic heat exchangers.

The generic form of the heat exchangers that we have fabricated is a hollow metal structure with faces connected by a network of cylinders that are 250–400 μm in diameter, or by a network of stripes that are ~750-μm wide. They have thin walls 100–300 μm to allow rapid heat transfer from the heat source to the cooling fluid. Our results only demonstrate the process and the types of features that can be fabricated; they do not represent optimal (or even realistic) designs. Because the fabrication process can be adapted to many types of structures with minimal change, it should be directly applicable to structures with performance much better than those described here. The performance of the nickel heat exchangers that we fabricated is, however, comparable to the performance reported for copper modules made by micromachining tools.

II. EXPERIMENTAL

A. Materials and Equipment

SU-8 photoresist (Microchem, Inc.; Newton, MA), tridecafluoro-1, 1, 2, 2-tetrahydrooctyl-1-trichlorosilane (United Chemical Technologies Inc., Bristol, PA), PDMS prepolymer (SYLGARD 184; Dow Corning Corporation; Midland, MI), polyurethane (Norland Products, Inc., New Brunswick, NJ), epoxy prepolymer (Epoxy Technology, Billerica, MA), granular PMMA (Average molecular weight = 996 000; Aldrich; Milwaukee, WI), tetrabutylammonium fluoride (TBAF) (1 M) in tetrahydrofuran (Aldrich, Milwaukee, WI), nickel sulfamate electroplating solution (Technic, Inc., Cranston, RI), and copper P electroplating solution (Technic, Inc., Cranston, RI) were all used as received. A mercury UV lamp (Hanovia) was used to cure PU or epoxy. A laser-sighted radiation thermometer (IR100, Dwyer Instruments, Inc., Michigan City, IN) was used to determine the temperature of the heat source during tests of the performance of the heat exchangers. Two adjustable water pumps (DUETTO DJ50 and MINI-JET MN 404, Aquarium Systems, Mentor, OH) were used to control the water flow through the heat exchanger. A pressure tester (PT14, Anderson Manufacturing Company, Inc., St. Paul, MN) was used to measure the pressure of the water flowing through the heat exchangers.

B. Fabrication of Heat Exchangers

We first used a computer aided design (CAD) program to draw patterns for the mandrels. We printed these designs on a transparency film using a high-resolution (3387 dpi) printer. [6]
This transparency was used as the mask for photolithography to pattern SU-8 photoresist on a silicon wafer. SU-8 films 100 microns thick required an exposure time of 35 s under UV light with a power density of 10 W cm\(^{-2}\). The time of exposure scaled linearly with increasing thickness of the SU-8. The patterned SU-8 was silanized with tridecafluoro-1,1,2,2-tetrahydrooctyl)-1-trichlorosilane and filled with PDMS prepolymer. The PDMS structure was cured by heating it at 70°C for 2 h. After curing the PDMS mandril, we removed it from the master manually, by peeling it away. Release is not a problem. For polymer mandrils from PU (or epoxy), we cured PDMS prepolymer around the PDMS mandril to make a negative replica of it as a mold. We then filled the mold with prepolymer of PU (or epoxy) and cured them by UV irradiation for 2 h before separating the mold from the mandril for the FMMA mandril. We cured the PDMS at 70°C for 2 h. The mandril was separated manually from the mold. Because PDMS has a very low surface free energy and because it is an elastomer, it is very easily separated from even complex molds. We used shadow evaporation or sputtering to deposit thin layers of chromium or gold on the mandrils. The sample was held at an angle of 30° relative to the source in the thermal evaporator to deposit chromium onto one side of the polymer mandril, then the mandril was flipped for the deposition onto another side. We used the layer of Cr or Au as the cathode to electrolyte nickel or copper to a thickness of 100–300 μm onto the mandrils. Electroplating proceeded at a rate of ~100 μm/day. The metallic heat exchangers were produced by dissolving PDMS in a 1 M solution of tetrabutylammonium fluoride (TBAF) in tetrahydrofuran (THF) or by burning out the PU, epoxy, or PMMA at 400°C in air.

C. Characterization of Heat Exchangers

A resistive heater composed of an array of gold wires (210-μm wide, 200-nm thick, 7-mm long) separated by 100 μm and connected to gold contact pads ~1.5 mm wide was used as a heat source. The gold heater was prepared on a silicon wafer using lift-off photolithography. [7] The wires of our heat source have a width of 0.21 mm, a length of 7 mm, and separation of 0.1 mm. The circuit also includes two gold pads (1.5 mm × 10 mm to make electrical contact with the gold wires. The total area of the heat source is 1 cm\(^2\). A thin layer of gallium was used for thermal contact. [7] The gallium film was prepared by spreading a drop of the metal (m.p. = 29.8°C) between the heat exchanger and the gold heater. The two structures were soldered with the gallium layer at room temperature. The heat exchanger with attached wafer was wrapped with an insulating aluminum oxide tape to avoid the loss of heat by radiative cooling and by convective cooling by air.

III. RESULTS AND DISCUSSION

A. Thermal Modules Fabricated Using Soft Lithography Techniques

Fig. 1 illustrates the process used to fabricate the nickel heat exchangers. It involves using soft lithography to fabricate a sacrificial polymer mandril, evaporating a thin film of chromium onto the polymer surface, and electroplating nickel onto the chromium layer to form the metallic microstructure. The polymer mandril was removed (by dissolving in an appropriate solution or by burning out in air) to yield the final heat exchanger. The key component in the fabrication process is the sacrificial polymer madril. We have used two different strategies, both based on soft lithography [1], [2] to generate these madrils. Replica molding [8]–[11] [Fig. 1(a), (b)] was used to fabricate PDMS, PU or epoxy mandrils, and microembossing [12]–[14] was used to fabricate PMMA mandrils.

When making mandrils out of PDMS [Fig. 1(a)], we first spin coated a 300–700-μm-thick layer of SU-8 photoresist on a silicon wafer, and patterned the SU-8 by photolithography; aspect ratios ranging from 0.5 to 3 were achieved by this process. We filled the mold with PDMS and cured the polymer at 70°C for 2 h. The mandril was separated manually from the mold. Because PDMS has a very low surface free energy and because it is an elastomer, it is very easily separated from even complex molds. We next used two sequential vapor depositions to coat the mandril with a thin (20 nm) layer of chromium on both sides; this layer of chromium acted as cathode for subsequent electrodeposition. To form the nickel layer, we used a commercial nickel sulfamate electroplating solution to electroplate nickel onto the PDMS membrane. In order to minimize the warping of PDMS in the electroplating solution, the PDMS structure was attached.
Fig. 2. Optical micrographs of nickel heat exchangers prepared by electrodeposition on polymer mandrils. a) Thermal module fabricated using a PDMS membrane (channel size 300 μm and cylindrical features 500 μm). b) Thermal module fabricated using a PU membrane (channel size 300 μm, stripe width 750 μm, and stripe separation 500 μm). c) Thermal module fabricated using an epoxy mandril (channel size 300 μm and cylindrical features 500 μm). d) Thermal module fabricated using an epoxy mandril (channel size 500 μm and cylindrical features 250 μm). e) Thermal module fabricated using an epoxy mandril (channel size 300 and 150 μm posts); f) Structure 2c operating at a water flow rate of 4.8 L h⁻¹.

to a glass slide by a binder clip prior to the nickel electrodeposition. The glass support was detached (by releasing the clip) once the membrane became stiff—that is, when the electroplated nickel films reached a thickness of approximately 50 μm. The electroplating process was continued until the thickness of the nickel layer was about 100 μm. To obtain the final heat exchanger, we dissolved the PDMS in a solution of TBAF (1 M) in THF for 4 h at room temperature[15], [16].

The procedure using PDMS works smoothly, but avoiding distortion due to the flexibility of this elastomer requires care. When a more rigid structure is required, one can also work with rigid polymers [Fig. 1(b)]. In this variant of the procedure, we used PU or epoxy to make the sacrificial mandrels. Because it is difficult to remove the rigid PU or epoxy mandrels from a rigid mold of SU-8, we used molds made from PDMS when working with these polymers; the PDMS molds were made from a negative replica of the PDMS mandrel. We filled the PDMS mold with PU (or epoxy) prepolymer, and cured the prepolymer by irradiation with UV light for 2 h. Once cured, the PU (or epoxy) structures were separated manually from the PDMS molds. We coated the PU (or epoxy) mandrels with chromium and electroplated nickel by the same procedure as that used for PDMS mandrels. The solid polymers are difficult to dissolve, so we removed them by heating the structure at 400 °C in air to obtain the free-standing heat exchanger.

Since most commercial polymers are available in solid form and are cheaper than liquid prepolymers, we developed a process that uses granular polymers for making the mandrels [Fig. 1(c)]. To demonstrate the process, we fabricated nickel heat exchangers using PMMA mandrels. The PMMA structures were prepared by microembossing: [17] PDMS molds were filled with granular PMMA, heated at 300 °C for 2 h to melt the PMMA particles, and cooled to room temperature. We detached the PMMA mandrels manually from the PDMS molds, and used the procedures described for PU (an initial coating of chromium followed by electrodeposition of nickel) to fabricate metallic microstructures. We tried to electrodeposit nickel on PMMA mandrels that had been sputtered with gold, but the gold layer flaked off the PMMA mandrels during the electrodeposition process. This flaking may be due to the thermal expansion of the PMMA, and to the poor adhesion of gold to PMMA.

We also used the processes in Fig. 1 to fabricate heat exchangers whose walls were composed of copper (100-μm thick) coated with nickel (50-μm thick). The nickel coating reinforces the structures. The copper layers were deposited using a commercial copper P electroplating solution. In our limited experience with copper structures lacking the nickel reinforcement coatings, the copper thermal modules bulged due to the high gas pressures generated inside the specimens when the mandrels were burned out or degraded in a THF solution of TBAF. For production of undistorted metallic shells using this variant of the procedure, it will be necessary to improve the method of removing the polymeric core.

Fig. 2 shows optical images of several nickel heat exchangers that we fabricated using mandrels made from different polymers. Structure 2a was made by electroplating on a PDMS membrane with a thickness of 300 μm. We designed this device to have an array of unfilled cylinders (400 μm in diameter) and a macroscale pentagonal void by the outlet. Structure 2b was prepared using a PU membrane with a thickness of 300 μm. This exchanger was designed to have stripes that were 750-μm wide and separated by 500 μm. Structure 2c was constructed by electrodeposition on an epoxy membrane with a thickness of 300 μm and with unfilled cylinders (400 μm in diameter) in the structure. Structure 2d was fabricated using an epoxy membrane with a thickness of 700 μm and with unfilled cylinders (250 μm...
Fig. 3. Illustration of the system used to study the thermal properties of the heat exchangers. a) Representation of a gold circuit generated on a silicon wafer (wire width 210 \( \mu \text{m} \), wire thickness 200 nm, wire separation 100 \( \mu \text{m} \), and wire length 7 mm). b) Representation of the setup employed for the heat dissipation tests.

A thermal module (see structure 2e) with nickel posts was also fabricated using an epoxy membrane (300 \( \mu \text{m} \) thick) and with holes (150 \( \mu \text{m} \) in diameter). The holes were totally filled with nickel when the thickness of metal film came close to 300 \( \mu \text{m} \). Fig. 2(f) shows structure 2c operating at a flow rate of water of 4.8 l h\(^{-1}\).

B. Performance of the Thermal Modules

We examined the performance of the nickel heat exchangers by measuring their thermal resistances (\( \theta \)). Fig. 3 shows the experimental setup that we used. A gold resistive heater (Fig. 3(a)) was placed on top of the heat exchangers, and heat was generated resistively by passing current through two copper wires that had been soldered (60Sn/40Pb alloy) to the contact pads of the gold heater (Fig. 3(b)). The change in the temperature of the gold heater was measured as a function of the electrical power applied to it and as a function of the flow rate of coolant passing through it. A similar system has been used by Nayak et al. to study the thermal properties of copper heat exchangers.[7]

Fig. 4 summarizes the performance of the nickel heat exchangers 2b, 2c, and 2e. For comparison, the temperature variation of a gold heater without a heat exchanger is also included in Fig. 4(a). The thermal resistance and maximum dissipation powers measured for these thermal modules are presented in Table I. The thermal resistances at a flow rate of water of \( \approx 20 \) L. h\(^{-1}\) indicate that the thermal module with posts 150 \( \mu \text{m} \) in diameter (2e; \( \theta = 0.07 ^\circ \text{C} \cdot \text{W}^{-1} \cdot \text{cm}^{-2} \)) is more effective than that with unfilled cylinders 400 \( \mu \text{m} \) in diameter (2c; \( \theta = 0.10 ^\circ \text{C} \cdot \text{W}^{-1} \cdot \text{cm}^{-2} \)). The module with stripes (2b; \( \theta = 0.14 ^\circ \text{C} \cdot \text{W}^{-1} \cdot \text{cm}^{-2} \)) is the least efficient. The performance of these thermal modules is also illustrated in the plots of thermal resistance vs. flow rate of water (Fig. 5). Theory [5] predicts that a plot of the thermal resistance of a heat exchanger vs. the inverse of flow rate should yield a straight line \( \theta \propto \frac{1}{(\rho C_p f)} \), where \( \theta = \text{Thermal resistance} \), \( \rho = \text{Density of coolant} \), \( C_p = \text{Heat capacity} \), \( f = \text{Flow rate} \), \( C_p = 4.18 \text{ J} \cdot ^\circ \text{C}^{-1} \cdot \text{cm}^{-3} \text{when water is used as coolant} \).

\[
\theta \approx \frac{1}{(\rho C_p f)}.
\]
TABLE 1

<table>
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<tr>
<th>Module</th>
<th>Flow rate (L h⁻¹)</th>
<th>Dissipation (W cm⁻²)</th>
<th>Tₐ (°C)</th>
<th>Tₛ (°C)</th>
<th>Thermal resistance (°C W⁻¹ cm⁻²)</th>
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<tr>
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</table>

* Temperature of water at the inlet.
*b Temperature of the heat source.

Fig. 5. A graph for the thermal resistance vs. the inverse of water flow rate corresponding to the nickel heat exchangers 2b, 2c, and 2e.

At a flow rate of water of ~20 L h⁻¹, the pressure of water flowing through structure 2b was 8.3 × 10⁴ N m⁻², that for 2c was 8.6 × 10³ N m⁻², and that for 2e was 7.1 × 10⁴ N m⁻². We believe that the thermal module 2c showed the lowest flow resistance because all the cylindrical posts are clustered at the center of the structure and it presents a low-resistance path (and a part that will lead to low thermal efficiency) at the periphery. Structures 2b and 2e can withstand pressures ≤2.1 × 10⁵ N m⁻² at flows ≤10 L h⁻¹ without leaking cooling fluid.

The nickel heat exchangers 2b, 2c, and 2e perform better than the copper heat exchangers reported by Nayak et al., which were fabricated by machining copper channels on blocks and soldering them to copper plates. The channels of these copper devices were 5870-μm wide, 1000- or 100-μm deep, and separated by 2500 μm. The thermal resistances for a copper exchanger with channels 1000-μm deep was 0.43 °C W⁻¹ cm⁻² (water flow =680.4 L h⁻¹), and that for one with channels 100-μm deep was 0.46 °C W⁻¹ cm⁻² (water flow =68.4 L h⁻¹). [7] Thermal resistances of the thermal modules 2b, 2c, and 2e are also comparable to the values measured by Tuckerman and Pease for their silicon heat exchangers (0.09 °C W⁻¹ cm⁻² at a water flow of 31 L h⁻¹). [5] Their structures consisted of silicon wafers with grooves that were 50-μm wide and 300-μm deep. The channels were closed by anodically bonding a Pyrex cover plate to the etched wafers. Using this design, they removed 790 W cm⁻² from a heat source while its temperature increased 71 °C above ambient. The high water pressure across Tuckerman and Pease’s design, 2.1 × 10⁵ N m⁻², is a shortcoming of this design because of the power consumption required to force water at useful velocities through the narrow channels.

Several groups have improved the performance of the heat exchangers designed by Tuckerman and Pease by modifying the geometrical configuration of the thermal modules. For instance, Mahalingam prepared a silicon heat exchanger with channels that were 200-μm wide, 1700-μm deep, and separated by 100 μm. This device had thermal resistances of 0.03 °C W⁻¹ cm⁻² (flow rate =43.2 L h⁻¹) or 0.02 °C W⁻¹ cm⁻² (flow rates =226.8 L h⁻¹). [3] Missagga et al. also extended the work of Tuckerman and Pease by alternating the direction of coolant flow in adjacent channels to obtain heat exchangers with uniform temperatures (±0.1 °C) across the heat source. They prepared devices that had thermal resistances of 0.14 (flow rate =56.9 L h⁻¹). Water pressure =7.3 × 10⁴ N m⁻² and 0.11 °C W⁻¹ cm⁻² (flow rate =100.8 L h⁻¹). Water pressure =2.5 × 10⁵ N m⁻²). [4]. Generally, the temperature of unidirectional flow heat exchangers, including our structures, varies by ±2 °C across the heat source surface.

IV. CONCLUSIONS

The procedure presented here can be used to construct a wide variety of unitary metal heat exchangers. The idea of using electrodeposition on sacrificial mandrils is, of course, well explored. [18], [19]. What is useful here is the demonstration that soft lithography provides a practical method for fabricating these mandrils with features that have dimensions of ~100–200 μm. With modest development based on exclusive previous experience with micromolding using soft lithography, we believe it will be possible to fabricate substantially smaller and more complicated features [20]–[23].

The polymers used to construct the heat exchanger should be selected according to the design of the device. For instance, it is difficult to avoid the bending of free-standing PDMS sacrificial membranes in an electroplating solution under vigorous agitation. A PDMS mandril with long, linear features [e.g., the design in Fig. 2(b)] can also fail by collapse and pairing of these features. The temperature of the metal evaporator where the thin PU and epoxy mandrils (≤300 μm) are coated with chromium should not be higher than 50 °C in order to avoid undesirable thermal deformations in the polymer mandrils. Electroless deposition of the cathode layer may circumvent these problems. The elasticity of the PDMS membranes, and the high temperature malleability of the PU or epoxy structures, can be desirable for the fabrication of nonplanar or three-dimensional heat exchangers by twisting, wrapping, or stretching initially planar polymer mandrils into more complex shapes prior to electrodeposition.
Although the performance of our prototypes is comparable to that of the most efficient thermal modules reported in the literature, the optimum heat exchanger configuration and the maximum thermal dissipation possible for exchangers of the types described here, remain to be established.

We believe that the microfabrication of metallic structures by electrodeposition on sacrificial polymer layers prepared using soft lithographic techniques has many potential applications in the fields of microelectromechanical systems (MEMS) and microfluidics. [24] Aside from heat exchangers, it should be feasible to fabricate microheaters, combustors, catalytic reactors, and also to fabricate a range of useful components such as flow devices and filters.

REFERENCES


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