Fabrication of Microchannel Chemical Reactors Using a Metal Lamination Process

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Abstract

Incorporation of microchannel arrays into small-scale chemical processing devices offers significant advantages in improving thermal control in the reaction region. Development of methods to produce high aspect ratio microchannels within solid metallic structures is an important step in the evolution of man-portable and other small-scale chemical processing units requiring high temperature and/or high pressure capabilities. Microchannel arrays can be used to facilitate heat removal or addition at the reaction zone or can be used to pre-heat gases prior to reaction. Staff at the Pacific Northwest National Laboratory (PNNL) have developed a method for producing solid metal components incorporating complex microchannel arrays by using a lamination and diffusion bonding process. This method uses metal shims that have been machined such that, when properly stacked, alternating microchannels and fins are produced in the laminated structure. The widths of the microchannels are determined by the thickness of the shim material, and heights and lengths of the channels are determined by the machined areas on the shims forming the channels. Machining of the shims can be accomplished in large quantities at low cost per shim by using photochemical machining or stamping processes. Consolidation of the laminated stack into a solid, leak-tight metal device is accomplished at elevated temperature and pressure by diffusion bonding. Microchannel arrays formed using this process can be produced either on the interior of the finished device or on an exterior surface. Typical microchannel dimensions in chemical processing devices produced at PNNL are 250 microns wide by 5000 microns deep. Application of the lamination process combined with a sheet-flow architecture can be used to produce highly compact chemical processing units. Among the devices produced at PNNL using this method are catalytic fuel processors and fuel vaporizers. Examples of all-metal stainless steel microchannel chemical processing devices produced using the lamination/diffusion bonding process will be presented and discussed.

Keywords: microchannel, chemical reactor, lamination, combustor

1 Introduction

Small-scale chemical processing devices are receiving increasing interest for military, automotive, and remote applications where size and weight limitations are an important consideration. Typical applications include heating and/or cooling devices, fuel processors, and chemical synthesis units [1,2]. Such small-scale devices provide unique design challenges that are inherent to the scale to which they are limited. For example, heat transfer mechanisms must be an integral part of the design of any chemical processing device in which heat is either released or absorbed by the reactions involved. In conventional-scale devices, this is generally accomplished through some form of heat exchange mechanism involving a heat transfer fluid confined within secondary containment and that flows either into or around the reaction zone. Thermal management in chemical processing devices intended for man-portable or other similar-scale applications can be particularly challenging as a result of the need to maintain highly compact, lightweight designs while simultaneously accommodating the materials requirements for high temperature and/or high pressure operation.

Staff at the Pacific Northwest National Laboratory (PNNL) have developed a method for fabricating all-metal small-scale chemical processing units using a lamination process [3,4]. These devices are produced from patterned metal shims that, when assembled, form complex features embedded in the internal structure of the final product. The stacked shims are diffusion bonded under high temperature and pressure, yielding a monolithic metal block. This method allows the production of solid, leaktight metal devices containing integral internal microchannel flowpaths for reactants, products, and heat exchange fluids. Thus, issues related to the use of gaskets or other such sealing devices used in high temperature/high pressure devices can be essentially eliminated. Because all liquids and gases involved in the process can be confined within the as-built device, fluid interconnects are also minimized.

An additional important benefit of the laminate fabrication method can be found in the fact that it allows the formation and incorporation of unique microchannel arrays and headers within the completed device without the need for additional machining. For the purposes of this discussion, we define a microchannel as having at least one dimension (typically the width) <250 µm. Arrays of parallel microchannels separated by thermally conductive fins provides an efficient mechanism by which to transfer heat either to or from fluids moving through the microchannels. Thus, microchannels can be used to promote enhanced heat transfer within the device at locations where heating or cooling is necessary or desirable. We believe that the fabrication method described provides a unique approach to the production of solid metal chemical processing units containing built-in thermal control and heat transfer mechanisms.

We describe the general procedures used at PNNL for fabricating laminated stainless steel devices for small-scale chemical processing applications. Specific

considerations related to the use of this fabrication method are discussed, and examples of stainless steel units fabricated to date are presented.

2 Microchannel Device Fabrication

In the simplest configuration, parallel arrays of microchannels can be formed by alternately stacking thin shims containing channel features with solid shims acting as fins separating the channels. The elongated channels in the channel shims are machined or etched through the shim so that the channel area is typically open. The pattern for the fin shims contains holes aligned with either end of the channel ends of the channel shims. When the channel and fin shims are alternately stacked, a series of parallel channels are formed that are bounded on either side by solid fins. A header is formed at either end of the channels by the holes in the fin shims that are aligned with the ends of the open channels in the channel shims. Widths of microchannels produced by this method are determined by the thickness of the shim material used to produce the channel shims. Shim thicknesses used for producing the types of devices discussed here typically range from 25 to 250 µm. The microchannel heights and lengths are determined by the patterned area of the channel shims. Using this assembly method, microchannels having a wide range of aspect ratios (height/width) can be produced. Typically, endplates having sufficient thickness to accommodate fluid connections are added to either end of the stack of patterned shims to complete the laminated device.

The design for a simple heat exchanger device consisting of two adjacent series of parallel microchannels is shown in Figure 1a. In this illustration, a hot fluid is directed through the lower series of channels. Heat is conducted through the fins between adjacent channels and into the thin layer of solid material separating the two levels of channels. It is then similarly conducted into the upper set of fins and into a cooler fluid flowing through the upper channels. Typically the intervening layer of solid material is kept as thin as possible to facilitate heat transfer. In an alternative design (Fig. 1b), two fluids can be made to transfer heat directly through the fins by flowing the hot and cold fluids through alternating channels in the microchannel array. In this design the headers for the fluids are located in the corners of the microchannels and alternating channel shims are flipped 180° to allow access of the respective fluids only to alternating channels. Using this type of design, heat transfer need only occur across the thickness of the fin shim, reducing heat transfer resistance by minimizing the conduction pathway. Flows of hot and cold fluids in this type of microchannel device can be configured in either a co-flow or counter-flow mode, as required for a specific heat transfer application.

Production of solid metal microchannel devices such as those described here can be broken up into several distinct steps, each with its own considerations. In summary, those steps can be grouped into the following categories:

1. device design
2. shim patterning
3. shim cleaning and assembly
4. bonding
5. post-bonding processes (optional)

We discuss each of these steps in more detail below.

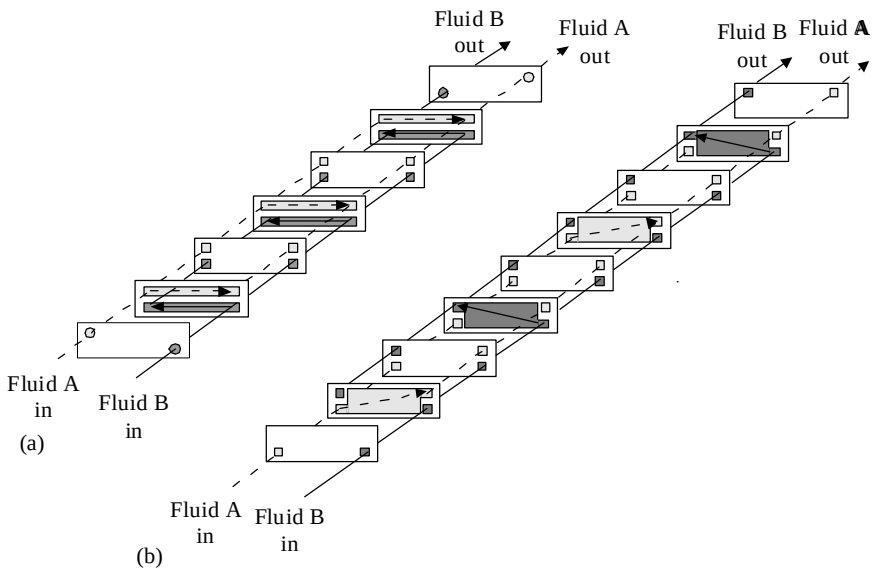


Figure 1. a) Design for a simple heat exchanger device; b) alternative design

2.1 Device Design

Generally, microchannel devices to be fabricated by the lamination process contain a series of parallel fluid flowpaths at different levels within the device. The sets of microchannels are vertically aligned and heat is transferred between fluids flowing in adjacent levels. The designs of the individual shims making up the device must be considered carefully to allow ingress and egress of fluids to the levels desired. At the same time, care must be taken to ensure that, when stacked into the final configuration, the shims will allow uniform transfer of pressure from one side of the stack to the other on all surfaces that require bonding.

For some applications, laminated devices may need to be built as separate components (e.g., to facilitate catalyst loading or replacement). Sealing issues between the components must be addressed using o-rings or other sealing

mechanisms. Sealing surfaces generally need to be machined after the bonding process to achieve an adequate finish.

2.2 Shim Patterning

For proof-of-principle and development work, we have found that the individual shims used to fabricate stainless steel laminated devices can be produced at relatively low cost (<$1 US/shim) using a photochemical etching process. In larger quantities, the cost/shim could be reduced by at least a factor of two using this process. Photochemical etching produces shims having acceptable tolerances in the thicknesses of material (50 to 500 µm) and given the flowpath dimensions commonly employed (0.1 – 1.0 cm). This patterning process has the capability to produce shims having highly complex patterns with no surface burring. Other patterning processes such as laser machining or stamping may also prove viable for producing shims for specialized applications or in mass production.

The endblocks used to sandwich the stacked shims and provide fluid interconnects are machined on a per-piece basis. This process could be automated for producing a laminated device in large quantities.

2.3 Shim Cleaning and Assembly

Patterned shims are vapor degreased prior to assembly to remove residual photoresist from the patterning process and any other organic contaminants. Prototype devices produced by this fabrication method have been hand-stacked, but this process could be automated large-scale production.

2.4 Bonding

Stacked shim/endblock assemblies are bonded into a single solid piece under vacuum using a high temperature/high pressure diffusion bonding process. Laminated parts produced to this point have been bonded by stacking the endblocks and shims into a pre-oxidized high temperature alloy clamping device to provide alignment and side support. Bonding is accomplished in a vacuum hot press. An alloy endplate and ram extension are used to transmit pressure from the hot press ram to the stacked laminate. A set of conditions that has been used to bond laminated stainless steel devices such as these described here is 920°C and 4000 psi for 4 hr.

Alternative diffusion bonding processes are currently being explored that would allow bonding to be accomplished in a vacuum furnace without the need for an external ram. The modified process uses the difference in thermal expansion coefficients between the clamping device and the laminated material

to produce the pressure required for bonding at elevated temperature. Ultrasonic bonding processes are also being evaluated for producing laminated devices.

2.5 Post-Bonding Processes

Because the bonded laminate acts as a solid piece of stainless steel, additional machining may be performed on the laminated parts after bonding. Such machining may be necessary to achieve smooth surfaces for o-ring or gasket seals, to provide additional fluid inlet or outlet ports, to reduce the overall mass of the resulting part, or for aesthetics.

Stainless steel tubing is commonly brazed or welded onto the fluid inlet and outlet ports of the laminated part after bonding. This connection method takes up much less surface area on the finished part and requires less material depth than other connection methods such as machined fittings.

3 Laminated Microchannel Devices

3.1 Microchannel Catalytic Combustor/Reactor

A small-scale combustor/reactor was designed to provide man-portable production of hydrogen for proton exchange membrane (PEM) fuel cells. The device was designed with a catalytic reactor section to process hydrocarbon feedstocks (methane, alcohols, gasoline, etc) into partial oxidation products, and a combustor section to pre-heat reactant gases. An all-metal design was stipulated to allow operation at temperatures approaching 1000°C in the combustion zone. Although not necessary from a fabrication standpoint, the device was produced in three sections to allow loading and replacement of foam metal catalyst supports. Two of the three parts comprising the device were fabricated using the lamination method to incorporate microchannels for enhanced heat transfer. The third component was a solid stainless steel plate required to seal the top of the device.

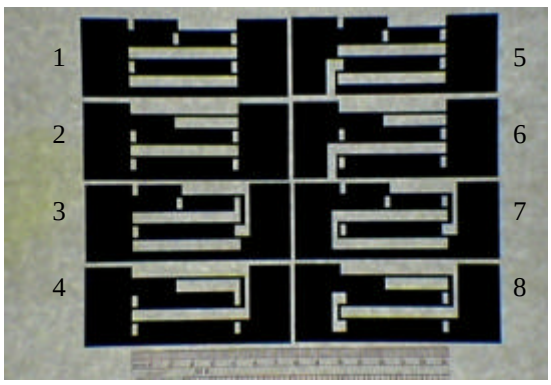
Patterned shims used to produce the laminated combustor/reactor are shown in Figure 2. All shims were photochemically etched in 250 µm-thick type 316 stainless steel. Eight different shim patterns (Fig. 2a) were required to produce the reactor body in order to accommodate the various coolant, fuel, and gas product flow paths required in this section of the device. The assembly order for the reactor part of the device was:

1. solid end block
2. 12 each shim #3 and 13 each shim #4, alternating
3. 26 each, shims #1 and #2, alternating
4. 8 each, shims #5 and #6, alternating

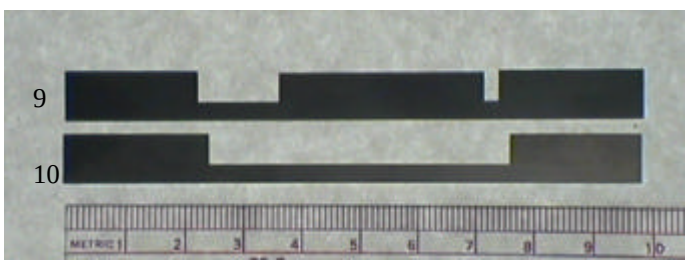
5. 26 each, shims #1 and #2, alternating
6. 12 each shim #7 and 13 each shim #8, alternating
7. endblock with coolant outlet

The combustor section of the device consisted of only two shim designs (Fig. 2b) and had no internal flow channels when assembled. The assembly order for the combustor was:

1. solid end block
2. 85 each shims #9 and #10, alternating
3. solid end block



a)



b)

Figure 2. Shim designs used for fabrication of microchannel combustor/reactor. a) Shim designs for reactor sections. b) Shim designs for combustor section.

After diffusion bonding at the conditions outlined in the previous section, the components were machined to add grooves for "C"-cross section metal seal rings between the components and a series of bolt holes around the perimeter of the device. Excess metal was also machined from the corners to reduce the weight and thermal mass of the finished device. Stainless steel tubing was welded onto the device at all inlet and outlet ports to facilitate fluid connections. A

photograph of the three parts of the completed combustor/reactor is shown in Figure 3.

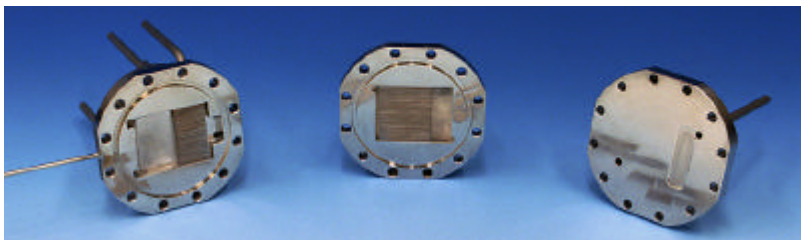


Figure 3. Three components of the laminated microchannel combustor/reactor

3.2 Microchannel Gasoline Vaporizer

Another device fabricated using the laminate process was the microchannel gasoline vaporizer, the first stage within a fuel processor to be used for automotive applications. As with the combustor/reactor described above, the gasoline vaporizer was designed with a catalytic combustion zone using an auxiliary fuel to provide the heat required to drive the primary function of the device. Microchannel heat exchangers were used to effectively transfer combustion heat to its area of use.

The gasoline vaporizer was produced using only two shim designs (Fig. 4) and two solid end plates. The shims were designed such that, when alternately stacked, two parallel series of microchannels were formed, separated by 1.7 mm of solid metal. Similarly, all fluid headers were produced by the design of the stacked shims. A total of 268 shims were used to produce the device. When bonded, the device formed open cavities designed to hold metal foam-supported combustion catalyst. After bonding, excess metal was removed to achieve a final shape. A groove was machined to accommodate a metal “C”-section seal ring, bolt holes were added around the perimeter, and ports were added for the liquid gasoline inlet, vaporized gasoline outlet, and the reaction gas exhaust. A solid top plate containing the inlet port for combustion gas was also fabricated. The flow diagram for the completed gasoline vaporizer is shown in Figure 5. Hot product gas from the reaction zone was directed into a series of microchannels on the level below the combustion zone before being removed through exhaust ports in the sides of the device. Heat was transmitted through fins of the microchannels and into the gasoline being pumped through the series of channels below. The microchannel gasoline vaporizer was demonstrated at full-scale for an automotive application, vaporizing 265 cc/min of gasoline.

4 Summary

A lamination/bonding procedure was developed for producing novel all metal small-scale chemical processing devices capable of operating at high

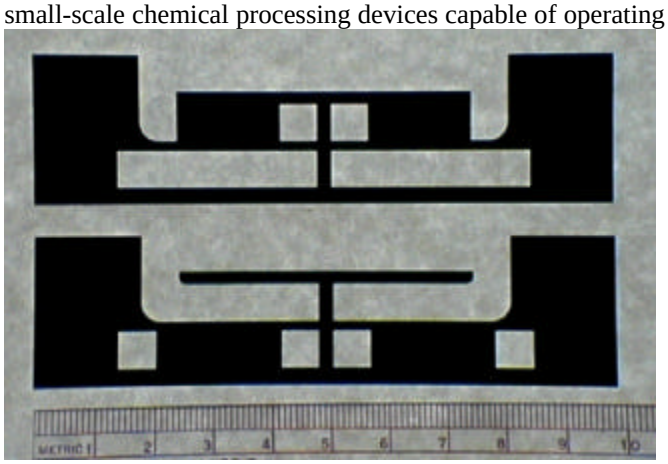


Figure 4. Shim designs used to fabricate the laminated microchannel gasoline vaporizer.

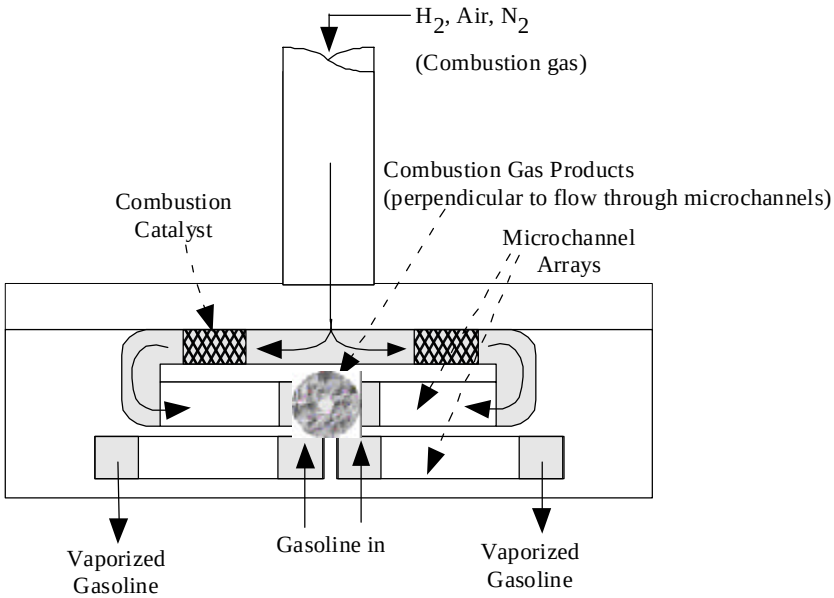


Figure 5. Flow diagram for laminated microchannel fuel vaporizer.

temperature/high pressure conditions. Such devices are suitable for portable or small-scale chemical processing applications. This fabrication method allows the construction of unique, highly compact, leak tight, devices containing integral microchannel arrays for thermal control and heat exchange processes. Microchannels and other fluidic features can be completely contained within the internal structure of the solid laminated component. The microchannel dimensions produced using the lamination process are determined by the machined pattern on the shims and on the thickness of the shims used in the construction of the device. Costs of fabricating the solid laminated devices can be reduced through volume production, the use of automated assembly methods, and improved bonding procedures.

Examples of laminated metal devices produced using the lamination/bonding method include a man-portable catalytic combustor/PO_x reactor and a fuel vaporizer for automotive applications. A number of additional laminated microchannel devices are currently in various stages of planning and development.

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