EUTECTIC DIFFUSION BRAZING PROCESS
FOR JOINING ALUMINUM LAMINAE
WITH MACRO- AND MICRO-
SCALE FEATURES

by

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Presented to the Faculty of the Graduate School of

The University of Texas at Arlington in Partial Fulfillment

of the Requirements

for the Degree of

DOCTOR OF PHILOSOPHY

THE UNIVERSITY OF TEXAS AT ARLINGTON

December 2013

Acknowledgements

I acknowledge The State of Texas, The Department of Energy, The Defense Advanced Research Projects Agency, Center for Renewable Energy Science and Technology at The University of Texas at Arlington for the project funding and facilities I had the pleasure to work with.

I acknowledge Dr. Richard Billo, Dr. John Priest, Dr. Brian Huff, and Dr. Erick Jones for their direct guidance, support, education, and the opportunities presented throughout my time at The University of Texas at Arlington.

I acknowledge Dr. Brian Dennis, Dr. Frederick MacDonnell, Dr. Norma Tacconi, Representative Joe Barton, for their foundation of work directly and indirectly contributing to my education.

I acknowledge Julie Estill, Kimetha Williams, Richard Zercher, Ann Hoang, Dr. Victoria Chen, Dr. Jay Rosenberger, Dr. Susan Ferreira, Dr. Jamie Rodgers, Dr. J. Royce Lummus, Dr. Herbert Corley, Dr. Sheik Imran, Dr. Don Liles, Dr. Bonnie Boardman, Howard Bailiff, Jimmy Hanhart, and Jimmy Williams for their impression upon my education, life lessons, stories, and their determination to see me succeed.

I acknowledge Joseph Billo, Sree Bhupathiraju, Panita Suebvisai, Matthew Oseng, Hung Cao, Rakesh Raj, Mary Campbell, Ryan Oliver, Zirun Zhang, John Dickson, Mel Hoshut, Arianne Padre, Smitha Malalur Nagaraja Rao, Praveen Pandojirao-Sunkojirao, Rabab Ali, Paul Dorasil, Jon Le, Kristina Antekelian, Samir Babar, and many more for their patience and friendship during my education.

I acknowledge Jeff, Sheilagh, John, and the rest of my family for their love and support.

July 23, 2013

Abstract

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Eutectic brazing occurs when two metals, such as aluminum and copper, react and diffuse together at a temperature lower than either material would melt separately. Commonly used in electronics manufacturing, this brazing method provides the basis for a new process of joining a laminated device with micro fluidic features. The proposed process overcomes the challenges of aluminum oxidation resulting from traditional diffusion bonding of aluminum laminae and does not require flux or caustic material pre-treatments. The process also protects surface geometries and avoids warpage, which is common with other higher-temperature joining processes such as welding.

This research provides a foundation for manufacturing multi-laminae, microfluidic, micro-feature devices in metals, especially aluminum. A reliable method of joining metal laminae could help make the manufacturing of these types of devices cost effective and reliable. These advantages will lead to faster rates of adoption of microfluidic micro-devices in industries such as pharmaceuticals, energy, food, and others.

This work combined laminae of aluminum alloy 6061 with copper foil interlayers to produce a milli-channel heat exchanger and reactor device. The devices contained overlapping coolant and reactant channel systems arranged perpendicularly within each lamina. Related work suggests that diffused joints are mechanically robust in contrast to the parent materials.

A laser micromachining center was used to trim the copper foil interlayers and a vacuum furnace hot press served to join laminae for the device. A reliable, leak-tight joining of the laminae occurred. Closed-loop argon testing at high operating temperatures verified the seal of the joint. The process repeatedly produced leak-tight, milli-channel heat-exchange and reactor devices.

The joining process is advantageous for joining complex milli- and micro-scale fluidic devices, such as a heat exchange and reactor device. Reliable joining takes place without application of fluxes, metal deposition, hazardous chemicals, or expensive cleanroom support.

This process will also benefit future microfluidics-related research and the integration of such technologies as arrayed milli-channel heat exchangers and reactor devices in industry. These devices enable modular scaling of production capacity, on-demand chemical processing, and higher purity products.

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Chapter 1

Introduction

Miniaturization is the “scaling-down” of devices. Miniaturization brings about affordability (higher product yields, lower cost, more efficient, smaller, faster etc.), new products (reactors, drugs, medical devices), and new services (newer-generation wireless communication). Every push for a smaller magnitude reveals new manufacturing problems that require advancements or likewise miniaturization in processing components.

Miniaturization in the electronics, chemical and bio-engineering industries brings about the emergence of new manufacturing technologies, new products, and new problems. With every stage of miniaturization, from micron to sub-nanoscale features, profound advances in technologies can improve the way people communicate and live.

Engineers must employ new geometry designs and advancements in materials to manage this problem. As these devices shrink and acquire more complicated features (e.g. dense multi-core silicon or the accelerating adaptation of lithium ion battery packs in automobiles), engineers will have to find unique ways to manage issues such as heat dissipation, higher pressures, etc.

The miniaturization of chemical processing and bioengineering in particular can create unique opportunities for advancement of knowledge and the development of new products and services. Miniaturizing chemical processes, using microfluidic devices, produces an opportunity to move away from batch processing and towards high speed on-demand processing. Microfluidic devices make possible the production of higher-purity products. A microfluidic device can deliver near-perfect blending, very short residence time, and on-demand production of chemical products. The microfluidic device may integrate various heating/cooling fluid circuits and elements, mixing geometries, foam-producing turbulence geometries, gas inlets, and electrical devices such as thermocouples.

MICROFLUIDICS

A microfluidic device forms its advantages due to the characteristics of fluid behavior at small scales. The small channels of microfluidic devices force molecules into smaller spaces, thus enabling intimate contact that may not occur in a vat. Due to the closer proximity, molecules in a chemical process do not experience laminar flow (they do not move past each other) and can produce products with lower residence times (time needed for an entire volume of reagents to complete a reaction).

A microfluidic device can also operate with zero moving parts, with the exception of a pump. Conventional external pumps can transition to use with microfluidic devices. An array of such microfluidic devices can eliminate the need for vats, mixers, cleaning equipment, and the associated manual labor needed to charge, clean, and operate those devices. These features contribute to the high-reliability of microfluidic devices.

Pharmaceutical researchers can use microfluidic devices to conduct exhaustive chemical assays with new drugs, drug interactions, chemical products, petrochemical blends, and more. With the use of computer-controlled pumps and various feedback sensors, researchers can automate these studies.

Pharmaceutical companies can realize the potential for micro-devices in the exploration for new drugs, expansive bioassays, developing genetically tailored medicine, and limited-run production. Pharmaceutical drug manufacturers can realize accelerated and improved testing of new drugs by performing many assays using smaller amounts of drugs in microfluidic device systems. As noted, microfluidic devices allow for high output purity and less waste. The device consumes feedstock only when a user needs a product. This feature allows pharmaceutical companies to conserve high-value or rare ingredients and samples.

The energy industry can use microfluidic devices as well. Microfluidic devices can produce a vast amount of experimental data used to optimize chemical production mixes. For example, an oil company may realize savings by re-optimizing a diesel production process that produces a product with less sulfur than what is federally mandated. Changing the process may save the company money and still meet federal emissions requirements. They can carry out many temperature and pressure varying experiments in microfluidic reactor devices.

LAMINATION

Laminated devices, constituted of machined laminae, or plates of material, joined by processes such as brazing can provide high-pressure and high-temperature compatibility. Those interested in high-temperature and high-pressure processes, such as hydrocarbon improvement, will find laminated devices appropriate for their use. Laminated devices, for now, provide a means to carry out high-mix, on-demand, and consistent production.

Lamination designs require an understanding of material properties and processes required to join the individual laminae without distorting the interior surface morphologies. One should make special considerations for removing fluxes and smut from the interior or the devices. Some designs may make it impossible to remove such materials due to complex interior geometries such as serpentine channels.

Main constituent materials may consist of metal alloys, plastic resins, and alumina. The machine bonds these constituent materials through dispensing adhesive, ejecting the constituent material as a heated plastic, or by using laser sintering to melt the materials.

Unfortunately, porosity, material bond weakness, and high expense present themselves as disadvantages for current additive machining technologies. Some microfluidic applications require the use of high-pressure and temperature. Current additive machining technologies simply cannot deliver devices suitable for these applications.

Sealing microfluidic devices for use with high-pressure and temperature requires a process with repeatability that produces a weld seam as strong as or stronger than the parent materials. The parent materials for these types of devices likely will include metals that resist oxidation, hydrogenation, and other various types of corrosion depending on the planned use of the product.

The problems facing microfluidic device adoption, beyond material costs, include the complexities of manufacturing such small devices and joining intricate components. The small features within these devices require consideration for manufacturing. Various machining and forming technologies exist to create surface morphologies in laminae. The problem we examine in this work regards joining these laminae while conserving the machined features.

This research explores the development of a method to join aluminum alloy laminae to produce a microfluidic reactor device for use with liquid fuels research. We explore contemporary metal joining methods and weigh their advantages and disadvantages. We then discuss the chosen joining method and then develop a process to apply it to the manufacturing of microfluidic reactor devices.

Chapter 2

Literature Review

Microfluidic devices can benefit industry by producing consistent product, on-demand, with lower residence times needed for production. As metal heat transfer devices are desired, there is a need to produce them “rapidly and inexpensively” [16].

Batch processes do not guarantee a perfect blend of components or a consistent residence time. Microfluidic devices can provide near-perfect blending, on-demand production, and a levelled residence time [21] [22]. When expanded production is desired, multiple microfluidic devices can perform in parallel, allowing operations to “scale-up” and transition away from batch processes completely [21] [22].

ENABLING TECHNOLOGY OF MICROFLUIDICS

Microfluidic devices rely on the microfluidic phenomena of fluid behavior in small-scale channels and cavities [8]. Microfluidic devices are known to, due to their high surface to volume ratios, enhance mass-transfer of reaction systems and decreasing residence time which is the time needed to complete a reaction [8]. Microfluidic devices experience a lack of laminar flow, in which molecules of reagents are forced together through channels, enhancing mass-transfer.

Microfluidic reactors can make use of surface geometries to control fluid flow, mixing, and introduction of gases. Paul describes such microfluidic systems, so-called “Microtechnology-based Energy and Chemical Systems (MECS),” as those devices fabricated in engineering materials [8]. The advantage of microfluidic systems is their small unit scale, production scalability in through arrays, which also enables portability of processes that would otherwise be too cumbersome to transport [8].

DEVICE APPLICATIONS

Microfluidic devices can support a range of uses from drug mixing to fuel manufacturing. A synthetic microchannel can mimic the environments to facilitate the study of cells, such as nerve-cell axon growth, to enable highly-arrayed pharmaceutical bioassays [8].

Applications of microfluidic systems range from biodiesel production, synthetic gas-to-liquid production, and others. Paul mentions a list of uses for high-heat microfluidic devices in durable materials such as stainless steel or metal-ceramic materials that includes "steam super-heating for driving turbines, hydrogen steam reforming, gasification of coals and heavy oils, flue gas desulphurization, …waste heat recovery and process heating, incineration…, domestic furnace waste recovery [heat], mobile engine heat recovery, solvent separation, microcombustion, artificial kidneys, and fuel reformers for fuel cells [8] [18].

Patented work by Meng suggests applications include computer cooling, radiators, refrigeration, water heating, and fuel cell heating and cooling [19]. NSF funded work to recover heat for thermoelectric conversion relies on microfluidic heat exchangers [14]. Work by BMW supports feasibility of recovering mobile engine heat recovery through the use of thermoelectric means in which it is suspected microchannels plays a role [15].

RELATED FUNDED RESEARCH

A 2010 NSF/DoE Partnership on Thermoelectric Devices for Vehicle Applications leveraged $9M to investigate exhaust gas heat recovery [14]. The Thermal Transport

Processes program considers heat transfer devices and applications of nanofluids for solar-energy [14].

Another high-profile research work entitled "Dissimilar Metal Joining for Micro-Scale Medical Devices" was developed at Colombia University in 2011. The work considered the joining of dissimilar bio-compatible materials. They developed a laser joining process for nickel-titanium shape-memory alloy and stainless steel 304 wires, using no filler material. The project was funded with an award of $359,379.00 from the National Science Foundation's Civil, Mechanical and Manufacturing Innovation (CMMI) organization in August 2011.

DEVICE FABRICATION

For the individual laminae, microchannels may be machined by means of compressive molding [16] roll-forming [19], or cut with endmills (conventional machining) [20]. Other methods for fabrication include photolithography, and additive machining. Pan lists other methods, including LIGA (Lithographie Galvanoformung Abformung), wet or dry etching, electro discharge machining (EDM), focused-ion etching (FIB), and micro-milling with conventional endmills [19].

ALUMINUM AND ALUMINUM OXIDE

In this research, aluminum alloy 6061 was used as the lamination substrate upon which microchannels were machined. The lamination process of 6061 aluminum alloy developed here had to overcome mainly the obstacle of aluminum oxide.

Aluminum enjoys extensive use due to abundance in the Earth's crust. The metal has a high range of use, having a high strength to weight ratio, and recycles easily. For these reasons, aluminum is likely to be a common material used in the construction of microfluidic reactor devices. The lightweight material can be alloyed to enhance chemical resistance and then formed, machined, and joined into extremely complex shapes.

The problem with laminating aluminum is a naturally occurring surface coating of aluminum oxide. The oxide coating is hard and has a high melting point of 2072 ºC which is much higher than 660 ºC, the melting point of aluminum [4]. The temperature and force needed to defeat the oxide film surpasses the thermal and structural capacity of solid aluminum alloy 6061.

While the oxide enjoys high strength, the interior aluminum alloy remains soft. When heated to a temperature or forced to a breaking point of aluminum oxide, the interior aluminum softens, loses rigidity, and is unable to conserve the machined microfeatures on the faces of the laminae.

High temperatures processes such as welding would likely destroy surface machined features. Low temperature joining processes that involve soldering would soften when the microfluidic device is heated to its operating temperature of 300 ºC.

We found that Al 6061 deformation was unlikely to occur with brazing. The brazing occurs at a temperature much lower than the softening point of the parent aluminum material. A vacuum environment could be used to prevent oxidation which would also reduce or eliminate the need for flux. By eliminating flux, complicated flux-removal work is also eliminated.

SURFACE PREPARATION

The intention of surface preparation is to enhance bonding quality of a metal adhesive bond [3]. Metal pre-treatments include degreasing with trichloroethylene (liquid and vapor), an aqueous NaOH solution, or 50% by volume nitric acid, followed by a water rinse [5].

ALUMINUM JOINING PROCESSES

Joining processes for metals (such as iron, aluminum, steel) include fasteners [28], soldering [29], friction stir-welding [17], adhesives [23], welding [20], ultrasonic welding [18], brazing [19], and others. Laminating microfluidic devices requires the development or modification of a traditional method in manufacturing.

Joining processes stem from two types of joining. Mechanical and metallurgical joining. Mechanical joining, including processes threaded fittings, soldering, stir-welding, adhesives, hook and loop, join materials through surface attachment, relying on friction. Metallurgical joining includes processes such as direct diffusion, welding, sintering, brazing, change the solutions of joining materials to bond at a chemical level, mixing at a molecular level, and requiring large amounts of energy.

Some applications require special surface preparations, environmental considerations, semiconductor processes, and additives to evolve a joint or create chemical stability during the joining process.

Metallurgical joining methods require treatments suited for the specific solutions (alloys) involved in the joining work. Typical for metallurgical joining is the need to remove oxidation, often with high-heat or friction, and to prevent oxidation during the joining process. For welding, specific solutions of consumable filler metal and flux are used to dissolve surface oxides, build a weld bead, and protect the weld bead during the cooling process. Welding companies have tailored electrodes to fit specific types of welding work in attempt to match the near unlimited amount metal alloys.

MECHANICAL JOINING

Adhesives

Adhesives for bonding aluminum and various substrates, such as composites and wood, exist in most manufacturing sectors. Automobile manufacturing has commonly relied on the light weight of aluminum alloys for bonding panels, hoods, engine blocks, suspension arms, and a many other automobile components.

The oxide on a metal surface is considered highly polar and favorable for adhesive bonding [3]. Adhesion provides the ability to produce a part from two dissimilar metals that distributes stresses in a way that is more uniform than with conventional mechanical fasteners [3]. Adhesives often require application-specific surface preparations, cleaning, and sometimes a specific surface morphology to ensure a reliable adhesive bond.

Soldering

The process is called soldering by the American Welding Society if the wetting metal temperature is below 427°C (800 °F). Soldering requires minimal tooling. Equipment consists of flux, soldering irons, vice, and the solder. [2]. Soldering often requires a rosin core to reduce the production of refractory oxides on the surface of the solder bead to ensure good flow and connection of the joint.

Soldering is typically carried out at 400 °C (752°F) and the name of the process typically applies to any joining process that occurs below 427 °C (800°F) [4]. “Soft soldering” occurs at less than 399°C (750°F) [2] .“Hard soldering” occurs at more than 399°C (750°F) [2]

Many difficult to reach crevices and corners make it difficult to use hand tools and conventional fasteners. For example, connecting electronic components in computers with conventional fasteners would require many small screws or clips and time that would likely cause the assembled part to be prohibitively expensive. [2] The application of soldered joints allows for higher speeds of assembly over screws. [2]

Many coatings and finishes of the parent material will resist degradation during a soldering process, compared to higher heat processes like welding and brazing [2].

For aluminum, while tin-lead solder can be utilized, aluminum-specific solders typically consist of tin, zinc, lead, and aluminum. The flux must be applied and the solder must be applied at the same time the oxides are removed as aluminum [2]

To prepare the surface of aluminum and its alloys, the aluminum must be heated until hot enough for lead-tin solder to melt when placed on the surface. A wire brush pushes the solder into the surface of the metal. This causes breaks in the oxide layer and allows the solder to wet the surface of the aluminum [2].

Stir welding

Stir-welding involves a sacrificial metal rod spun at high RPM into the seam between two metals, which may be dissimilar.

METALLURGICAL JOINING

Welding

Welding aluminum occurs at or above the melting point of 1220°C. Welding can destroy physical properties of aluminum and aluminum alloys at high temperatures [5].

Diffusion

Diffusion is when two materials mix into each other. Diffusion bonding occurs when two materials heated to a temperature nearing their softening points join while force is applied. The force causes particles in the materials to mix at a molecular or elemental level. Diffusion bonding, typically processed with a vacuum hot press (or vacuum furnace press), is effective for harder materials The process of solid-state diffusion requires some time and features must be pressed uniformly [18]

Diffusion bonding requires the heating of parent metals above solidus and below liquidus. At these temperatures, a force applied to the metals causes the softened solid solutions to diffuse into each other creating a single bonded product with no discernible weld line and bond characteristics as if the part were constructed of a single unit of material.

6061 Aluminum alloy softens at a lower temperature than surface oxidation. 6061 Aluminum alloy melts at 582 ºC, which is much lower than aluminum oxide's melting temperature of 2072 ºC. Pressing force cannot break the aluminum oxide to alloy 6061 aluminum alloy to diffuse. Direct diffusion is impossible with this soft alloy if the oxide layer is present.

Table 2-1 Melting Points of Aluminum, Copper, Oxides

Material	**Melting Point**
Aluminum Oxide	2072°C
Copper Oxide	1201°C
Copper	1085°C
Aluminum	660°C
Aluminum Alloy 6061	582°C

Brazing

Brazing aluminum occurs above 427°C (800°F) but below the melting point of 660°C (1220°F). Brazing is used to join materials without melting them [1]. Typically used for heat-treatable aluminum alloys. Brazing requires less heat than welding. Filler metal must be liquid above 450 degrees Celsius [1].There are 6 major methods of brazing: dip brazing, furnace brazing, induction brazing, infrared brazing, resistance brazing, and torch brazing [1]. Brazing is typically better for the strength of different materials and their expansion characteristics. Modern brazing allows for strong, leak-proof joints made quickly and inexpensively [4] The liquidus of the filler metal must be lower than the solidus of the base materials.

Bonds from brazing metal as thin as 12.5 micrometers typically exhibit the same or better physical strengths of the aluminum parent material as well as the same levels of corrosion resistance[4]. Brazing can be used for hard to reach joints in complex assemblies, such as the undulating surfaces of heat transfer devices [4]. Quenching after brazing is standard practice and makes heat treatment generally unnecessary [4]. A traditional brazing filler material for aluminum 6061 is Al-12Si [7].

Parts must be cleaned and protected by fluxing or a protective non-oxygen atmosphere to prevent oxidation [1]. Vacuum furnace brazing often eliminates the need for flux as vacuum prevents oxidation [1]. Vacuum brazing has found adoption in industrial areas where reactive materials, such as aluminum and copper, are joined [1].

For brazing, fluxes prevent refractory oxide formation on the brazing material and the joining material. Flux also encourages the flow of braze throughout the accessible surface of the part. Fluxes can also regulate braze flow at a desired rate depending on application and can be applied in different ways. Fluxes may be brushed on or sprayed prior to brazing or may sit upon the brazing solution in a dip-braze process. Detergents can remove unwanted flux from the joined part. In small parts, fluxes can be difficult to remove. In microfluidic applications, fluxes can affect the flow through the narrow channels. Flux is difficult to remove from devices with complex shapes and small openings, such as heat exchangers [6].

Honeycomb parts are as difficult to remove flux from in the same way that heat exchangers are due to small openings and difficult shapes [6]. Honeycomb structures without perforations are impossible to braze [6]. Some claim that it is not economically feasible to fabricate honeycomb panels by coating both sides with an interlayer metal such as copper, silver, gold, tin, etc.

Eutectic Brazing

A form of brazing called eutectic brazing, uses two metal alloys that produce refractory oxides solids, to break up the oxide coatings and reduce them into the structure of the joint. Eutectic brazing may be used to create joints that are nearly as strong as the parent metals [5].

Most refractory oxides, such as aluminum oxide, have high hardness and high melting temperatures. While two separate metals may have two high melting points, their contact and heat applied can motivate the generation of eutectic alloys. Using a thin layer of silver, copper, gold, tin, or zinc as an interlayer [5], a eutectic reaction can dissolve the oxides and lead to diffusion in the weld seam. Films may be applied using vacuum and electrolytic deposition methods [5]. Additionally, vapor deposition may be used. Briggs & Dajas suggest the use of films no thicker than 10-3 inches [5]. Larger films may result in the formation of brittle intermetallic alloys that may result in a weakened bond [5]. Foils may be used as interlayer materials [7] [8]. Shin et al brazed Aluminum 6061 together using copper as an interlayer. They refer to the process as diffusion brazing – eutectic reaction [7].

Metal pre-treatments before use include degreasing with trichloroethylene (liquid and vapor), aqueous NaOH solution, rinsing in water, cleaning with 50% by volume nitric acid, and rinsing again in water [5]. Aluminum oxide must be removed before brazing [6]. Without a flux, it is necessary to clean all bonding surfaces of aluminum oxide and to coat all bonding surfaces with metal interlayers, such as copper [6].

Heating the pieces together to a temperature between the eutectic melting point and the parent material melting point causes a bond [5]. The eutectic temperature only needs to be exceeded by 30-60°F to allow the eutectic alloy to diffuse into the parent metal [5]. Pieces can be joined in a partial vacuum or inert gas heated to the eutectic temperature [6].

Formation of eutectoids between the mating surfaces allows diffusion of the interlayer metal foil into the parent metals being bonded [5]. The eutectic alloys diffuse into the parent metals leaving a visually faint weld line between the parent materials [5]. The eutectic material occurs quickly once the proper temperature levels are reached, allowing the aluminum bonding to occur rapidly [5]. Time needed for a successful aluminum-eutectoid bond is between 1.5 to 10 minutes.

Some claim eutectic seals can remain strong after prolonged periods of operation and temperature cycling [5]. Machined tube eutectic bonds hold up to 2300 psi at 800°F before the machined tube, not the bond, failed [5]. Others claim that a bond with a coating of only silver, copper, gold, and tin, cannot sustain any significant stress [6]. Bonded machine tube specimens with nuclear fuel gas lasted 90 days at 1000°F, remaining leak-tight [5].

Mechanism: The Eutectic Reaction

While copper has a melting point of 1080 degrees Celsius, the joining of aluminum and copper together requires heating to a lower temperature in which a eutectic process occurs. The eutectic point, or lower melting point between binary alloys of copper and aluminum, exists at 548°C.

Table 2-2 Melting Points of Eutectic Materials and Aluminum

Material	**Type**	**Melting Point**
Aluminum Alloy 6061	Multi	582°C
Aluminum-Silicon	Binary	577°C
Aluminum-Silver	Binary	554°C
Aluminum-Copper	Binary	548°C
Aluminum-Gold	Binary	527°C
Aluminum-Copper-Silicon	Tertiary	524°C
Aluminum-Copper-Magnesium	Tertiary	451°C
Aluminum-Magnesium	Binary	450°C
Aluminum-Zinc	Binary	382°C
Aluminum-Tin	Binary	242°C

Table 2-3 Melting Points of Eutectic Materials of Copper and 6061 Aluminum Alloy

Material	**Type**	**Melting Point**
Aluminum Alloy 6061	Multi	582°C
Aluminum-Silicon	Binary	577°C
Aluminum-Copper	Binary	548°C
Aluminum-Copper-Silicon	Tertiary	524°C
Aluminum-Copper-Magnesium	Tertiary	451°C
Aluminum-Magnesium	Binary	450°C

Application of Eutectic Diffusion Brazing

A study by Wells et al was conducted using lap joints [6]. The lap joints were coated with 0.0002 inches of copper utilizing electro deposition, followed by a thin layer of tin [6]. Only one side of the mated part needs to be coated [6]. This work was heated to a eutectic tertiary (aluminum, copper, tin) temperature of 540°C [6]. The work was heated in partial-vacuum with inert gas to limit the growth of oxides [6].

According to Briggs & Dajas, high-pressure solid-state aluminum eutectic (with tin, zinc, gold, copper, and silver in the alloy) bonding with pressures between 30,000 and 80,000 psi were used fuel element fabrication [5]. The application requires bonds between aluminum parts to be strong and leak-tight. Briggs & Dajas suggest that many high-pressure and high-temperature processes for bonding aluminum members is not feasible for bonding thousands of parts [5].

Aluminum is commonly found as a cladding material for nuclear fuel elements [5]. Briggs and Dajas patented a method of joining aluminum parts in such a way that the joint would be as strong or better than the parent metals [5].Their method involved eutectic diffusing, bonding as a method to form fuel-rod ends to seal in UO2 gases. The bond regions would need to exhibit no brittle oxide of intermetallic compounds. They bonded aluminum nuclear fuel caps with a diameter of .3 inches with a wall thickness of 0.030 inches with swaged 0.297 inch outer diameter inset tube of the same material.

Microelectronics manufacturers also employ eutectic brazes for joining electronic Al/Cu connections [7]. The high cost of gold and copper make low cost alloys of aluminum extremely attractive when microelectronics manufacturers produce in the millions.

Chapter 3

Formulation of a Question

With this work, we ultimately want to answer the question: how does one hermetically bond metal laminae with micro-features to withstand high temperature and pressure?

SPECIFIC OBJECTIVES

This research builds on previous micro-fluidic device research. The objective was to produce an aluminum alloy 6061 microreactor device suitable for hydrogenation of synthetic gas. Several challenges regarding materials, processes, and design, would be met through trial and error and building upon the results of related research work from other fields of engineering to the production of this device.

The use of high-temperatures and high-pressures for handling hydrogen and upgrading synthetic gas or natural gas created challenges in material selection Specifically, the material selected for the device would have to be compatible with hydrogen. The material would need to be corrosive resistant and stable in the presence of high-temperature high-pressure hydrogenation of the synthetic gas feedstock.

Any joining materials for the device would also need to be hydrogen compatible as well as capable of resisting corrosion and fracturing under the pressure and temperature changes the device would likely undergo during reactions. The design of the device includes features machined into the surface of laminae that would make it difficult to remove excess joining material or flux.

Tasks to accomplish the objective involved investigation of possible processes to identify possible processes for experimentation. To test the experimental processes, a testing procedure would need to be developed. The candidate joining processes would be tried, tested, reviewed, and improved upon with each experimental cycle. The goal of the objective was to arrive at a process that produces a sealed laminated device to meet the needs of hydrogenating synthetic gas in the presence of a catalyst. Common engineering materials and processes compatible with existing machinery in our laboratory were favored due to commercial availability, machinability, and costs.

PROSPECTUS

It is desirable to survey aluminum bonding processes for aluminum 6061 lamination in hopes of identifying a method that effectively seals the microfluidic device. From adhesion to welding, each method has a particular way of not just joining the aluminum substrate, but also dealing with the challenging layer of naturally forming oxides surface film. I hope to find a gap in methodology and discover a new technique or application for aluminum 6061 bonding that is readily adaptable to manufacturing.

HYPOTHESIS

The combination of aluminum oxide and complex internal geometries of a laminated microfluidic reactor device creates a unique roadblock that must be overcome with an improved metal joining process. Such a process will need to produce a strong diffused bond between the parent material laminae that provides a secure and reliable seal to contain high-temperature and high-pressure operations in the presence of hydrogen and corrosive byproducts. Ideally, the device would be one solid unit without discernible welds.

Such a process may allow a simple single-heating processing step to laminate the entire device at once. A single heating process will avoid the need for additive machining, such as powdered titanium laser sintering, which at the time of this writing was not a compatible process for high-pressure applications such as ours due to issues of porosity (and as we would discover, unfavorable hydrogen interaction).

An acceptable process would also require minimal material investment, avoiding exotic metals and dangerous chemicals. Several additive machining processes require multiple passes of build-up layers of material. These additive processes require specialized adhesive binders, structural sacrificial temporary materials, and many passes to build up the construction of the designed device. Additive machining would require the need to remove additive material powders from the internal channels of the device, which is arguably impossible to do. Some additive machining processes, such as those for ABS plastic resin, require the use of chemicals to dissolve temporary structuring materials used to hold up the printed part.

An ideal process will combine a simple material processing technology with our current conventional machining capabilities and will produce a sealed microfluidic device to carry out hydrogenation of synthetic gas.

Chapter 4

Methodology and Research Approach

The objective was to develop an economical process to seal multiple aluminum laminate that can withstand high temperatures and high pressures and protect the machined micro features on the interior of the device. The lack of time and resources, long process times, and cost of materials prohibited using a formal design of experiments approach. Rather, a systematic investigative approach was used. In our approach, we looked for opportunities of improvement, often looking back to the literature and analyzing alternative parameters such as temperature, material thickness, surface morphology, and design features.

TASK 1 INVESTIGATE POSSIBLE PROCESSES

An extensive literature review was performed to find appropriate manufacturing and joining methods for laminated microfluidic devices in Chapter 2. Special emphasis was spent on the pioneering work of Paul et al., Shin et al., Chu De, and Shirzadi.

TASK 2 IDENTIFY A TESTING PROCEDURE

Before attempting the different experiments, the testing procedure needed to be settled.. The testing procedure needed to be repeatable, safe, and focused on exploiting any weaknesses in the seal. Weakness in the seal would lead to mixing among hot combustible gases, heated oil, and the air.

Both channel systems, coolant and reactant, would be pressurized with argon gas at the inlet while the outlet was capped off. Only one channel system was pressurized at a time, to reveal cross-circuit leakage. The device would first be submerged in water to check for obvious leaks. If not leaks were detected, the device would be dried off and then submerged again under pressure but this time in a bath of hot silicone oil. The hot silicone oil would bring the device temperature up to testing levels, between 275°C and 300°C, what was above the normally designed operating temperature. An argon tank pressurized the device to 150psi, and later 300psi, during tests through stainless steel tubing. Possible leaks could be visually monitored. The pressure gauge on the argon tank would also be monitored. A leak of argon in the silicone bath would produce foam at the top of the bath as well. This method was chosen as an alternative to testing the device with flammable gases, such as hydrogen or natural gas, to avoid creating the opportunity for a fire.

TASK 3 IDENTIFY PROCESSES FOR EXPERIMENTATION

The overall process involves stacking laminae and doing something to use or alter the state of the layer of aluminum oxide between the laminae. Simply, the device process for experimentation involved joining the entire microfluidic device at once and sending it off for leak testing.

Several joining methods were identified for experimentation. These methods include: the application of direct diffusion, adhesive bonding, and finally eutectic brazing. These joining methods were chosen due to perceived compatibility with the device assembly design and the already acquired tools and machinery of our laboratory.

The process of experimentation focuses on manipulating control variables such as heat, time, number of laminae, thickness of interlayer materials, surface roughness of bonding surfaces, cooling times, and others. Each joining method requires a slight change in the amount of variables.

TASK 4 RUN EXPERIMENTS TO ESTABLISH A GOOD SEAL

Several methods were evaluated for joining micro-devices. Of these methods, some were not feasible. Chapter 7 will describe the various methods and their results.

Experiments included:

- direct diffusion of titanium to titanium
- direct diffusion of aluminum 6061 to aluminum 6061
- direct diffusion of anodized aluminum 6061 to aluminum 6061
- metal-ceramic adhesive between laminae
- eutectic brazing of aluminum 6061 with evaporative copper vapor deposition interlayer
- eutectic brazing of aluminum 6061 with copper foil interlayer

TASK 5 DEVELOP PROPOSED PROCESS

From the results of the experiments, a proposed methodology for laminating a microfluidic device was developed. These tasks are repeated in cycled until the desired result emerges. We have reported in the next chapter on each of the revision s made to both process and design as we approached a final positive resulting process.

Chapter 5

Analysis

VERIFYING THE SEAL (TESTING)

The design of the microfluidic reactor carries the capacity of handling a range of temperatures and pressures to focus the production of specific hydrocarbon chain lengths, thus influencing the outgoing product from incoming synthetic gas and hydrogen. To control this reaction environment, a user can control the inlet pressures and flow for the hydrogen and synthetic gas in one channel system and control the temperature of the heating oil in the other channel system. The two channel systems must exist in close proximity, but must not interact. They must be separate and must not leak.

A leak could involve high-temperature and high-pressure combustible gas or oil and create a danger for the surrounding area. Possible locations for leaks to occur include between laminae to the exterior perimeter of the devices, from an interior channel to an alignment hole to the outside of the device, between laminae at the location of an inlet or outlet corridor, and at inlet and outlet opening between the device and a poorly fit nozzle. We would later find that leaks between laminae were particularly likely to happen when force during bonding was not distributed evenly above the surfaces to be bonded. At the softening temperatures of the aluminum alloy, when the copper – aluminum eutectic alloy product would form, the aluminum alloy fails to distribute force evenly across its surface to parts below. This was due to an overlap of the hole for the inlets above a critical bonding surface adjacent to the manifold areas on the ends of the microfluidic reactor.

The microfluidic reactor device would operate at a range of temperatures near 150°C and pressurized with hydrogen at a maximum pressure of 80psi. As a student would ultimately use the device to carry out experiments to demonstrate the capability of a university-developed catalyst, the device needed to operate in a safe and predictable manner. The between-laminae joints needed to be leak-free and durable, with factor of safety which we interpreted as the capability of containing a higher than normal operating pressure at a much higher temperature.

Verifying the seal required creating a similar operating environment to what the device would ultimately be exposed to. The testing environment operated the device at 160psi over the course of 3 hours at 300°C. The maximum operating pressure typically used was 80psi with a max temperature of 300°C. The idea behind this test suggested that higher testing pressures would exploit leaks that would go unnoticed at 80psi, but could still pose a risk for combustion. We need a way to observe leaks and correct processing and/or design parameters in a safe way.

By exposing the micro-fluid reactor device to the higher pressure we can expect to cause a weak seal to fall apart at the joint. We understand that failures near the joint in actual operation have the catastrophic potential. The device operating temperatures and pressures prevent an opportunity for combustion in combination with the volatile synthetic and hydrogen gases and their fuel products.

For our test, we submerged the micro-fluid reactor in a bath of hot silicone oil to heat the device. The temperature of the hot silicone oil rose to be 300°C. Visual evidence of bubbles indicates at least one leak in the device. Leaks appear through incomplete seals on the outside area of the device or as a stream emerging from the topside holes. The devices were kept at temperature and pressure for a minimum of 4 hours. A tank of argon gas supplied the device with 160psi of pressure.

Submerging the device in hot silicone oil presents an opportunity for injury due to splashing that can occur if a large leak occurs. To detect larger, more obvious leaks, a researcher first submerged the device in a bath of water and applied argon gas pressure to the device. Early failures released argon gas into the water bath causing harmless splashes instead of eruptions of burning-hot silicon oil. This also saves time from being wasted on waiting for the silicone oil to heat up and having to clean away residual oil if the specimen has a large leak detectable in water.

As the design and joining approaches improved, leaks became undetectable in water. The observer placed the device into the bath of hot silicone oil after allowing it to spend some time to dry in air. Water boils at a lower temperature than silicone oil. To prevent burns, one should remove any moisture to prevent dangerous splashing of the hot silicone oil. One should also wear protective clothing, gloves, and a face shield.

An observer monitored the device in anticipation of any gas release in the form of bubbles or streams. Small leaks could also form a foam on the surface of the silicone oil. Argon emissions would occur on the outer edges of the device as well as within the interior of the device, viewed as streams of bubbles rising from the inlet and/or outlet orifices not connected directly to the argon supply line.

Figure 5-1 Leak-testing a microreactor in silicone oil

The silicone bath temperature was monitored using a Fluke electronic device with a thermocouple attachment. Argon pressurized the device though stainless steel tubing and fittings.

This testing setup allowed us to quickly decide if a successful bond occurred. The temperature was run at and sometimes higher than what would normally be used. At 300°C, silicone oil tends to smoke, so we could not test any higher than 320°C in most cases without heavy smoking. We die use higher pressures though. While we aimed to test the devices at 160psi, we continued to have success at 300psi.

Several material and process changes occurred in this project. We will now take time to review those changes. After several adjustments in design, surface preparation, treatment parameterization, and the assembly method, several working prototypes made their way to the laboratory for testing, and we eventually produced successful bonds.

DIRECT DIFFUSION WITH TITANIUM

The original micro-fluid reactor device design presumed direct diffusion to join titanium laminae with micro-channel patterns machined through photo-lithography. Direct diffusion uses heat and force to join like materials with little to no discernible bond line. Ideally, direct machining produces a part with no evidence of previously existing in separate pieces.

Direct diffusion for titanium requires the use of vacuum to prevent oxidation at high temperatures. The vacuum environment prevents oxidation since no air meets the specimen. Complex geometries may be brazed or diffused with such a machine. Vacuum hot press usage requires time to “vacuum-down” the processing chamber, which some consider as a disadvantage. Failing to do so can result in damage to heating elements through oxidation.

Figure 5-2 AVS Vacuum hot press

We procured a vacuum hot press (also referred to as a vacuum furnace press) from AVS, Inc. (Ayer, MA) to process the parts. A vacuum hot press works by vacuuming out the air inside the chamber to create a “hard vacuum” and uses graphite elements to produce infrared radiation to heat the parts contained inside the unit. The vacuum furnace press allows the pressing of metals that would oxidize in the natural environment. For materials like aluminum, this is important is it a naturally occurring aluminum oxide quickly develops in nature and makes joining aluminum difficult.

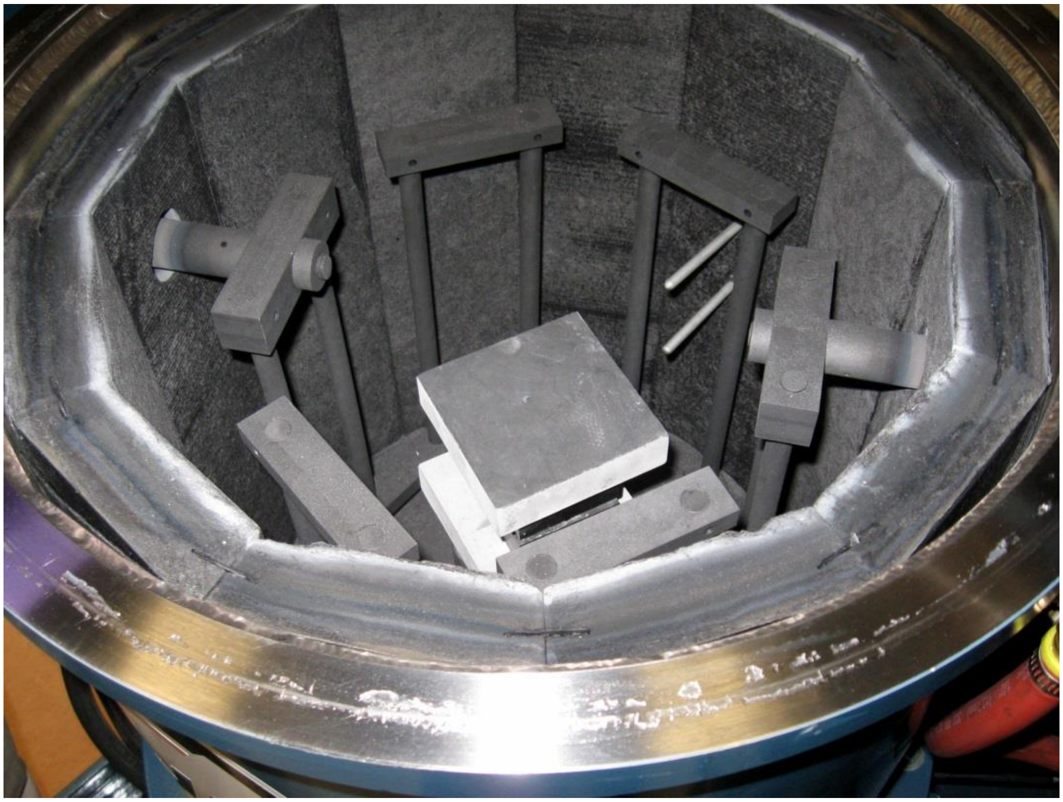

Figure 5-3 Interior view of the vacuum chamber of the vacuum hot press

The vacuum hot press processed the titanium laminae into a single micro-fluid device. The interior of the vacuum hot press is lined with graphite foam insulation. The heating elements are also graphite. The machine can process parts up to six inches cubed in size. A ram on the topside can apply up to five tons of force inside the chamber.

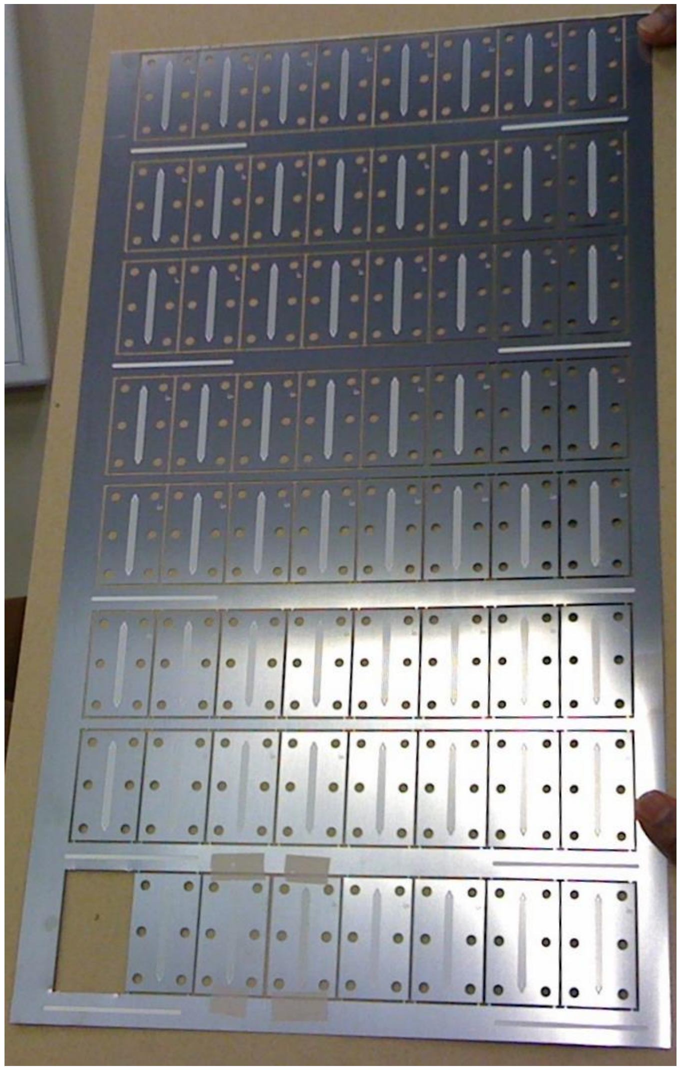

Figure 5-4 Titanium laminae produced commercially through photo-lithography

We acquired out titanium laminae in sheets produced by a company that specializes in photolithography of films and metals. The titanium laminae were uniform contained a set of channels in parallel on one surface.

We expect photolithography to be an appreciable method of producing vast amounts of identical metal laminae for the fabrication on mass-produced microfluidic devices in the future.

Figure 5-5 A specimen that received too much heat and force

Diffusion is a difficult process to work with. Due to naturally occurring metal oxides, diffusion of soft metals is unlikely to occur if an oxide layer is present. The oxide layer provides effective protection of the softer inner core metal. It is no wonder that anodizing of metals is a popular method of protecting and enhancing a product's appearance.

Oxide coatings behave similarly to candy-coated chocolates. As the interior becomes soft, the outer shell may crack and allow the interior material to flow outward and solidify when cooled. However, when dealing with lamination, the larger surface areas spread the force among the oxidized surfaces and prevents cracking from occurring. Instead, cracks will occur on the perimeter of the part, the part will stretch out, or the interior will flow out the sides.

We learned from these experiments that diffusion could only occur the oxide layer were to somehow vanish and the interior metals were softened only up until the point that they could deform.

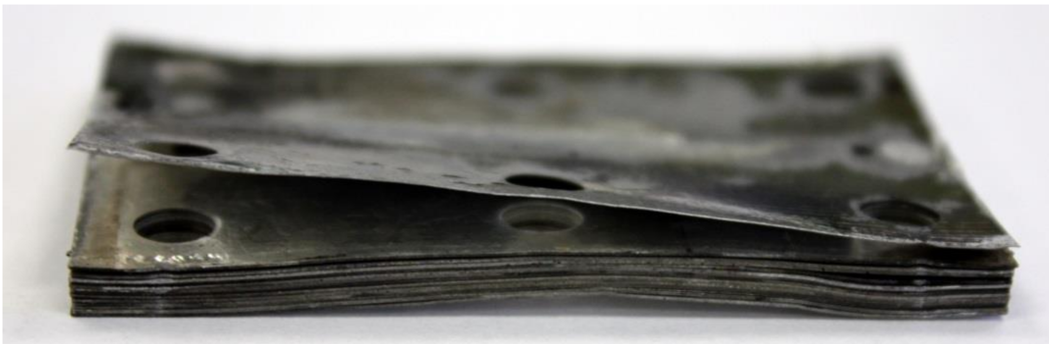

Figure 5-6 Oxidation also provides an obstacle for titanium direct diffusion.

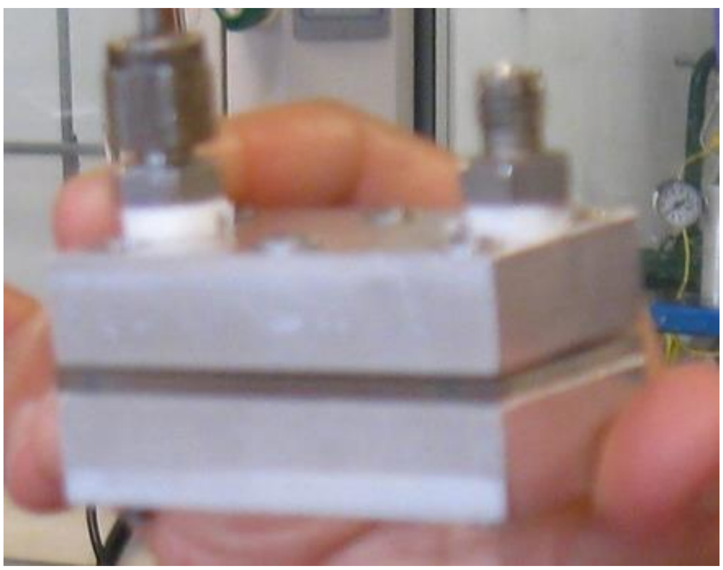

Figure 5-7 Titanium prototype device prior to hydrogen exposure.

The titanium laminae prototype device produced through direct diffusion in titanium did not survive exposure to the pressurized, hydrogen-rich, environment. According witnesses, the prototype device disintegrated due to hydrogen fatigue.

DIRECT DIFFUSION WITH ALUMINUM 6061

Following the titanium disintegration incident, a decision to change base material away from titanium brought about the use of aluminum alloy 6061. Aluminum alloy 6061 provides chemical stability in the presence of a hydrogen-rich environment. The alloy contains a mix of (in descending order of %weight, typically) aluminum, magnesium, silicon, iron, copper, chromium, zinc, and less than one-tenth of one percent of manganese, titanium, and other less substantial amounts of elements [10].

Computer-numerically-controlled (CNC) mills can machine the soft aluminum 6061 alloy with ease. This method of featuring does introduce some tension into the surfaces of the alloy due to its softness. Annealing can relax the surface tension.

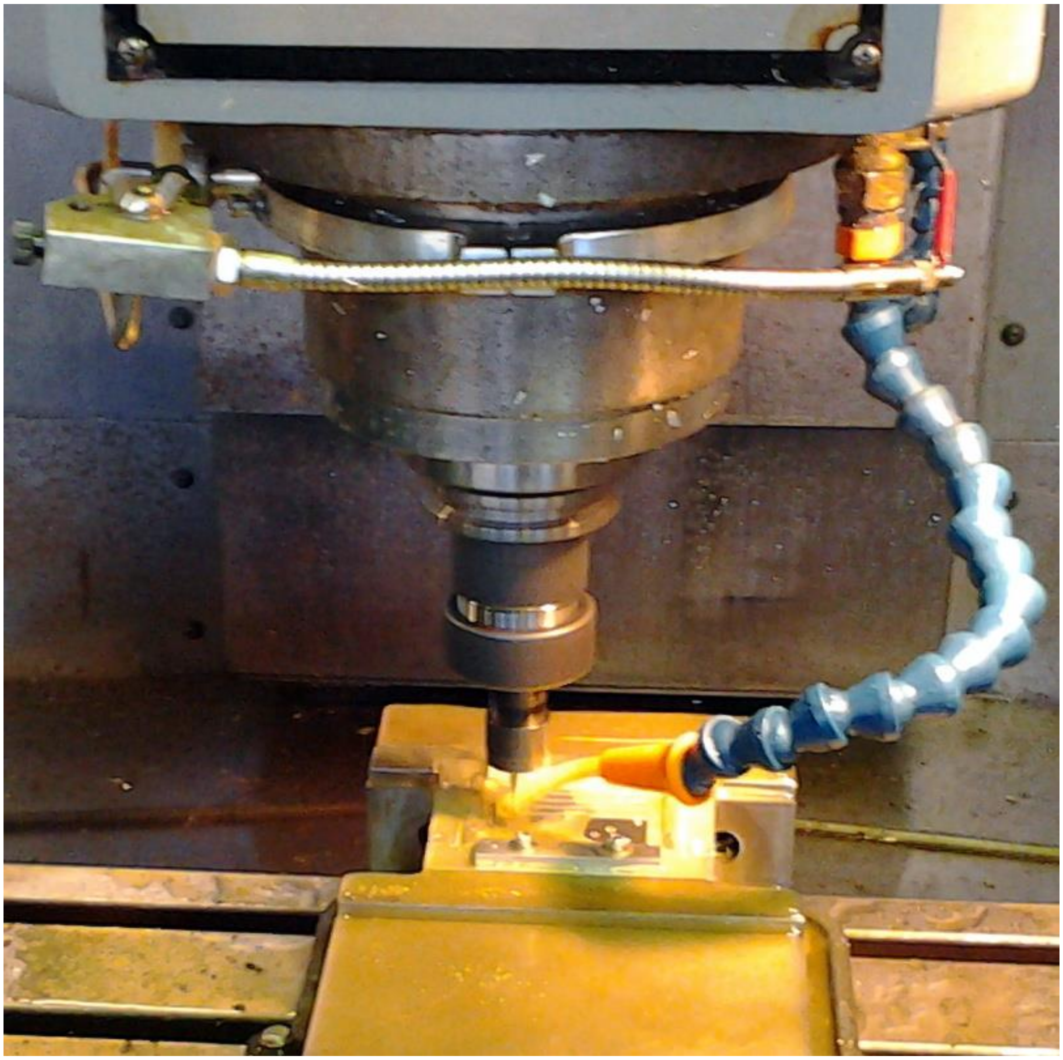

Figure 5-8 CNC milling of aluminum laminae

The 6061 alloy begins to soften at 582 °C and becomes a flowing liquid at 652 °C [10]. Aluminum oxide (Al_2O_3) melts at a significantly higher 2,072 °C.

Because of aluminum oxide hardness and inexpensiveness, industry has found many applications for it as an abrasive. One may find aluminum oxide in many polishing compounds, sand paper, and toothpaste.

Anodizing occurs when electrical current flows through a part, set up as the anode, to build up the oxide layer. We originally planned to apply this surface treatment to the inside of the aluminum laminae channels to attach a catalyst. One way would be to anodize the individual laminae and then bond them together. The alternative was to anodize the part from the inside out.

We wanted to know how the laminae would behave with the extra thick layer of oxidation provided by the anodizing treatment. This will allow us to visualize and understand the mechanisms behind how the aluminum oxide prevents the diffusion bonding process from occurring.

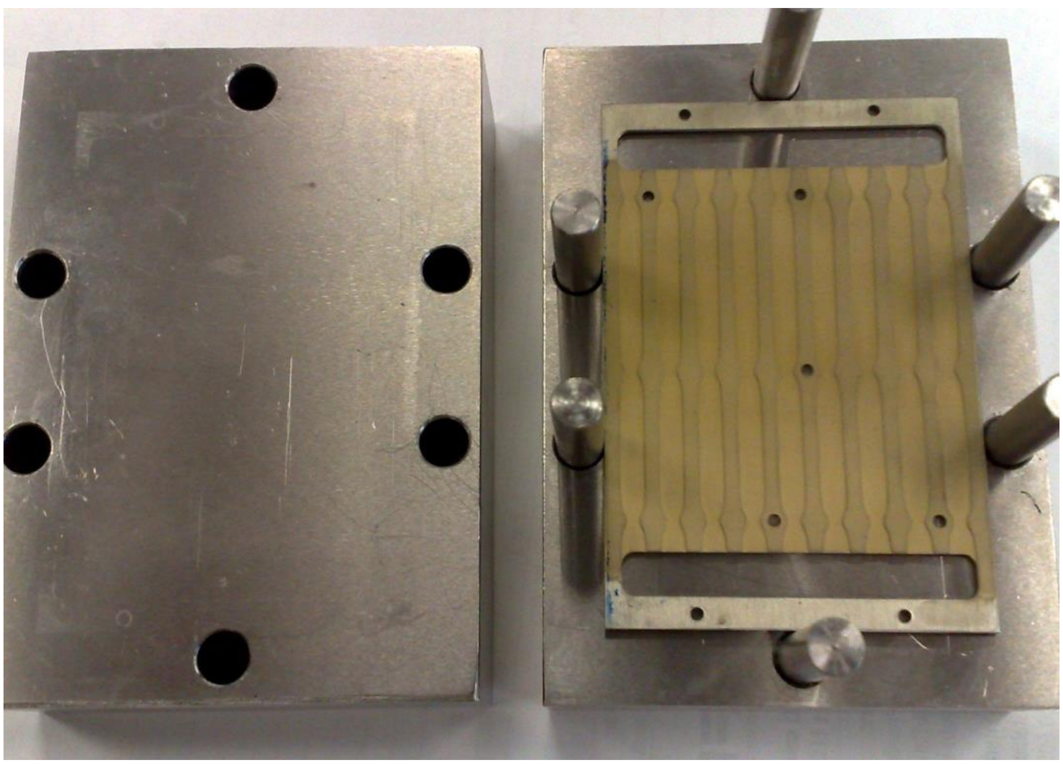

Figure 5-9 Anodized aluminum laminae prior to diffusion bond attempt

Aluminum oxide's hardness and high-temperature melting point make it a considerably difficult material to work with. Anodized aluminum exacerbates this feature.

Using direct diffusion, the temperature at which aluminum oxide would soften to permit diffusion of the aluminum parent material would exceed the solidus of the aluminum and cause the part to liquefy within the aluminum oxide shell. Features machined in the aluminum part would give way to the applied pressing force or the effect of gravity upon the now freely flowing aluminum alloy liquid.

Figure 5-10 Aluminum oxide prevents diffusion bonding

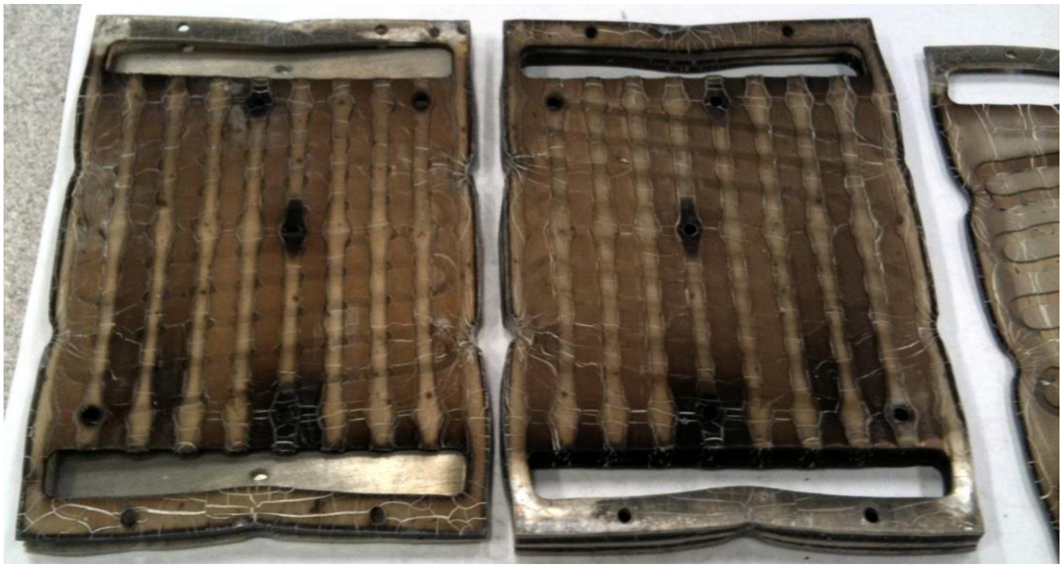

Figure 5-11 Aluminum oxide cracks as interior aluminum alloy deforms

We also tried to apply coatings of various enamel spray paints to explore masking the surfaces of the aluminum prior to anodizing. We wanted to see if we could introduce selectivity through masking the surface with an enamel coating. The anodizing process grows aluminum oxide, and we suspect the aluminum oxide crept underneath the coating, with help from the sulfuric acid used during anodizing, as it flaked off during the anodizing process.

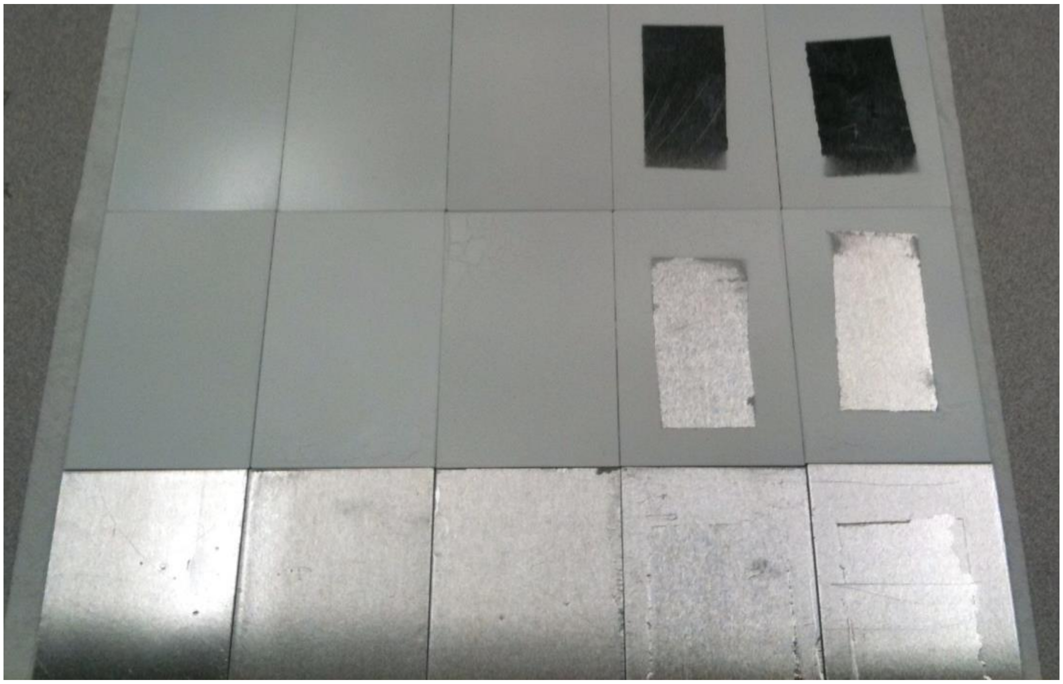

Figure 5-12 Aluminum test-laminae prior to anodizing with various spray-on coatings

We left attempting to anodize the aluminum parts to focus on purely bonding the aluminum parts and working to reduce oxidation as much as possible.

Without a flux to penetrate the aluminum oxide, or some chemical surface treatment combined with a zero-oxygen atmosphere to prevent oxidation, direct diffusion of plain aluminum alloy 6061 laminae might as well be considered impossible due to the enclosure of aluminum oxide that protects the parent aluminum alloy. Next, we considered using an adhesive to join the laminae.

ADHESIVE

An adhesive product supposedly compatible with our high temperatures, but not verified to be compatible with high-pressures of the micro-reactor, was tried. The product, Pyro-Putty 1000, manufactured by Aremco, could have been an ideal solution. The product was designed to be applied to cracks in furnaces or other high-heat machinery. It is unknown what types of pressures the product can accommodate.

The product comes as a two-part metal-ceramic powder paired with a curing solution that mixes into a paste (Powder 1000-P-1116, Liquid 1000-L-1116). The two need to be mixed 24 hours in advance of application in an airtight container (as per the directions). When applied, the paste can be heated to cure. The time for curing and the heat at which the product cures was not explicitly disclosed in the directions and likely is dependent on the powder to liquid ratio.

The company claims the product can be thinned to a desired viscosity and is described to contain inorganic resins. The product is described as a ceramic and aluminum two-part water-based paste. The product can be set in air or heat cured at 160°C. The company claims their product as stable up to 760°C. Applications include high-heat devices such as exchangers, boilers, and afterburners. The product is also used to repair cracks in cast aluminum parts.

We attempted to mix the product in vials and dispense it by syringe. To inject the product through syringe, we used a high amount of liquid. We tried three ratios: (liquid: powder) 1.5:2, 1.75:2, 2:2. We mixed the product 24 hours in advance in separate glass vials with airtight lids and left them to set up overnight. We attempted to use a syringe, but the mixtures were still too thick to be brought up through the needle. We instead used the needles as spatulas to apply the product which had a gel consistency. We used the needles to place the material on to the aluminum laminae. Then we placed the parts between clamps and allowed them to air drive for 4 hours. We had a second set sit in a hot press for 4 hours at 160°F.

We were unable to get the adhesive to activate as we had hoped.. The viscosity of the product was likely to blame. We spoke with Aremco associates and were told that we were adding too much liquid, still. So perhaps for other larger applications, a thicker putty would probably work. As for our application, we needed to apply very small amounts and the only feasible way to do this at the time with a syringe. A syringe, because of the small opening, would require high viscosity or high pressure.

Future work with metal-ceramic adhesives may look at using mechanically or digitally controlled adhesive applications. The adhesive is really more of a filler, designed to be applied as a patch that can be sanded and painted much like automobile plastic patch products. Perhaps a high-pressure dispensing system could move the paste.

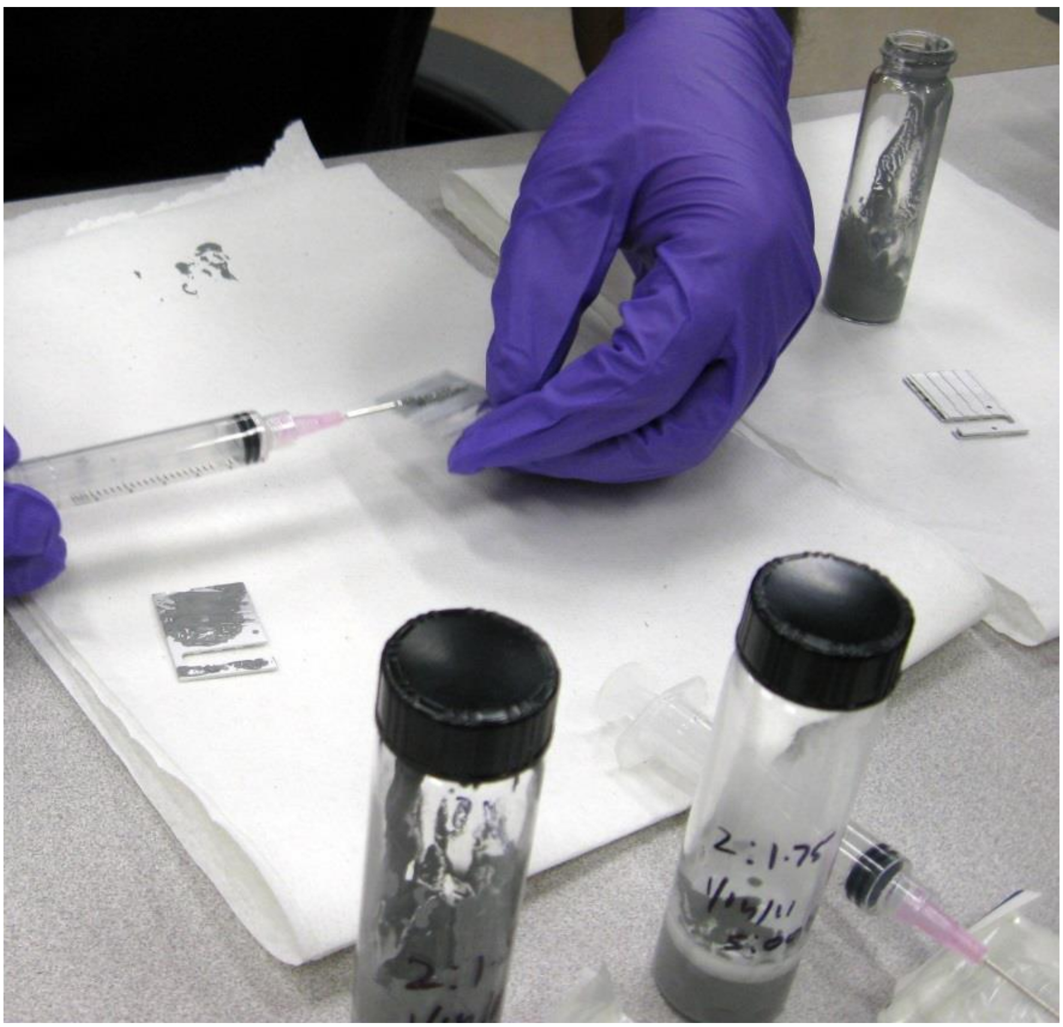

Figure 5-13 Attempt to coat with high-heat adhesive putty

We were unable to extract the Pyro-Putty into our syringes, so used them as spatulas to transfer the putty.

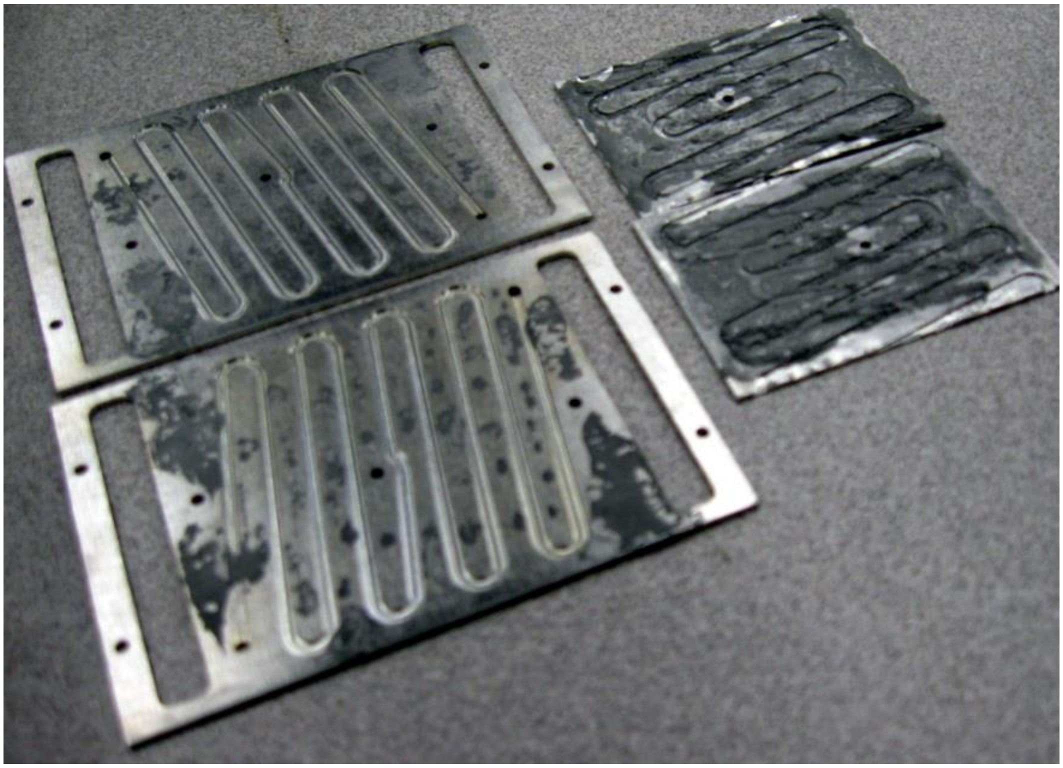

Figure 5-14 Attempt to coat laminae with high-heat adhesive putty

We were unable to spread the Pyro-Putty evenly on our small features. Future work may look into using different ratios of Pyro-Putty powder to liquid and identifying a better method of depositing the putty. If not by syringe, perhaps a stamping method or a brush could apply the adhesive.

EUTECTIC BRAZING WITH COPPER VAPOR DEPOSITION INTERLAYER

Literature review revealed previous work and industry standard practice of combining aluminum and copper wires for micro-electronics through the use of eutectic brazing. Aluminum is desired over copper due to its low cost.

The eutectic brazing process works by making use of the eutectic alloy that forms between copper and aluminum at a lower temperature than the aluminum and copper metals separately. The eutectic alloy creates a brazed bond between the two metals that is considered as good as or better in strength than the separate materials.

Machined laminae were coated with a vapor deposition layer of copper. This process was carried out in a vacuum chamber. Tungsten boats hold pure copper pellets in the bottom of the device and aluminum plates face the boat. The pure copper comes to boil, gasifies, and rises to coat the aluminum. The coating is about 250 nanometers thin.

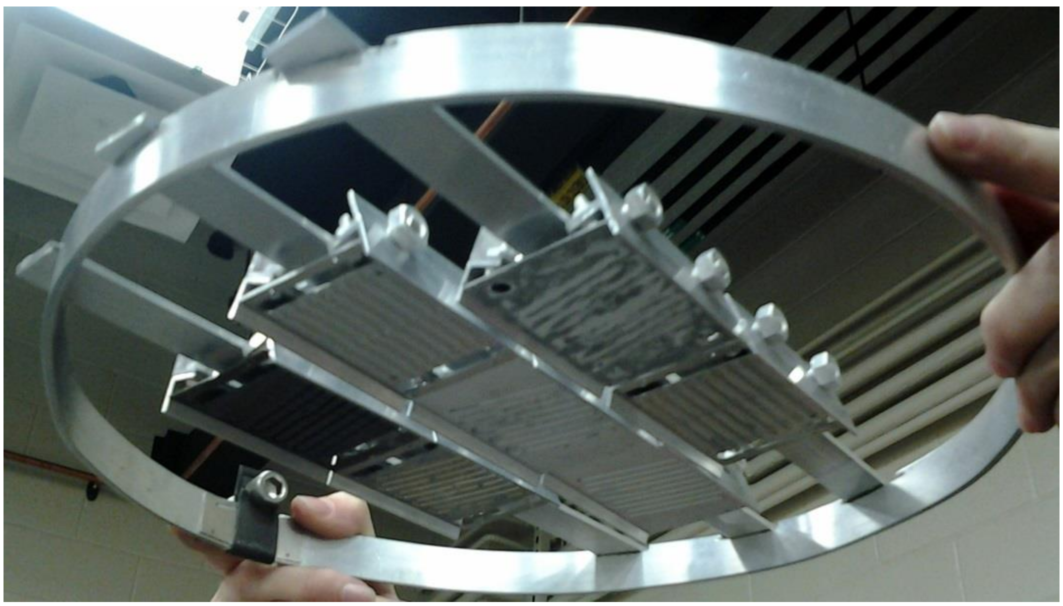

Figure 5-15 Fixture for copper vapor deposition upon aluminum laminae

These coated laminae were then stacked and heated. The part produced after processing in the vacuum hot press exhibited some sealing qualities, but still exhibited some leaks. The parts were heated to 550 C and held for 30 minutes.

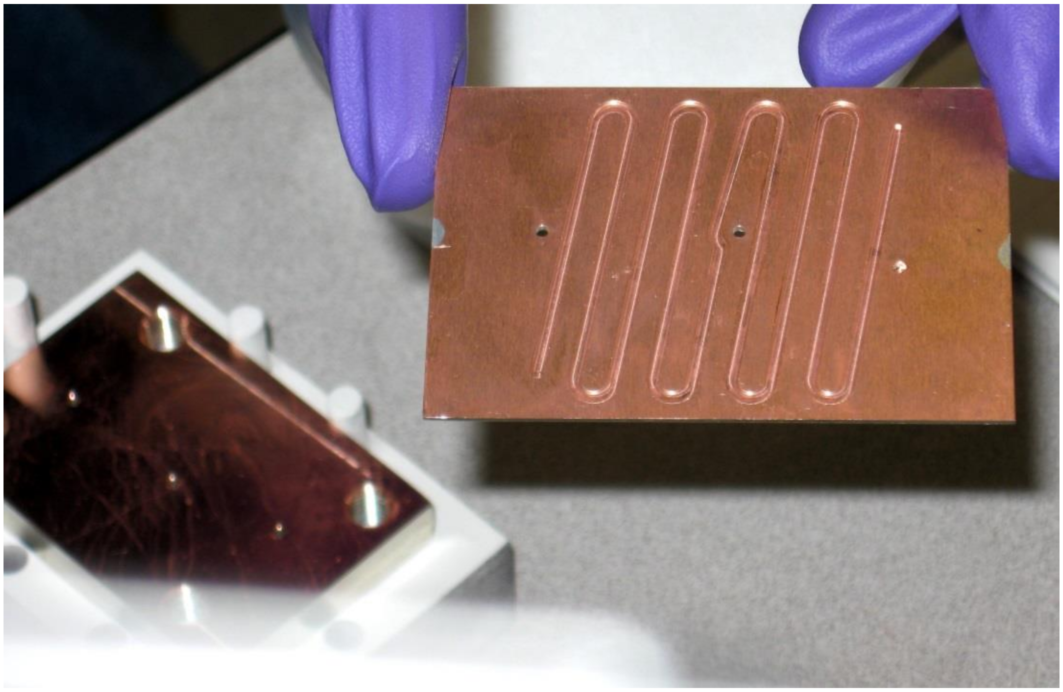

Figure 5-16 Copper vapor deposition onto the surfaces of the aluminum laminae

Although much smaller, we still noticed evidence of both external leaks and internal leaks. The external leaks were among the middle laminae and in seemingly random places along the perimeter of the part. The interior leaks could have been laminae-to-laminae, such as there may be a lack of seal between heating fluid and reactor channels or there could have been direct interior leaks from either the heating fluid or the reactor channels. We are unsure and currently unable to identify the location of this seal failure.

We decided to try a higher temperature (560°C) and longer residence times (40, then 45 minutes), but continued to produce leaking modules. We also added more time for copper deposition to allow a thicker coating of the copper vapor deposition. We were now coating with an estimated 600 nanometers of copper.

Continued increase of copper deposition contributed to less leaks and smaller leaks. This led us to think that much more copper was necessary to produce a leak-free seal. We found the eutectic process could produce more eutectic alloy braze if more copper was available on the surface of the aluminum laminae.

EUTECTIC BRAZING WITH COPPER FOIL INTERLAYER

We found a commercially available copper foil of 12.7 µm (0.0005 inches) manufactured by All Foils, Inc. (Cleveland, OH). Handling the foil requires some care as the foil can easily fold and wrinkle although the foil. The foil was thick enough to be easily handled by hand and was not brittle. The foil arrived in square pre-cut 6-inch by 6-inch sheets.

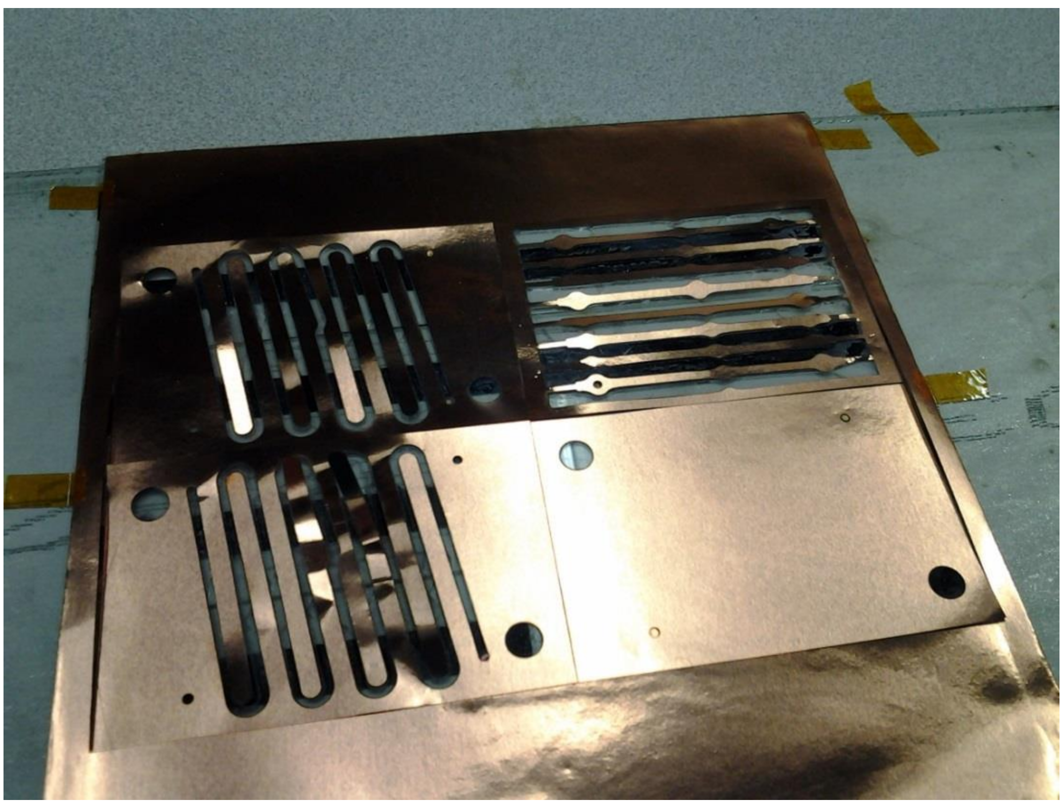

Figure 5-17 Copper foil interlayers after laser processing

We cut the copper foil using an Oxford Lasers (Oxfordshire, UK) Neodymium-doped yttrium aluminum garnet (Nd:YAG) frequency tripled laser. The 5W 350nm UV laser trimmed the copper foils into gaskets. The laser cut the copper gaskets upon a sheet of glass slides. We lifted the gaskets out from the cut sheets using Forceps. We used acetone to remove residual ablated materials (burrs, dust).

The process of cutting copper foil with a laser required taking a 3D model of the device and identifying the four unique gasket patterns needed to perform the eutectic bond. To do this, we reduced 3D models produced with Rhino to 2D drawings using Planit (now Vero Limited, Gloucestershire UK) Alphacam CAM profiling software. The geometries needed for the gaskets were isolated and traced into tool paths. The program converted the tool paths into numerical control (NC) code for the laser.

Four unique gasket designs were required. One would be below the thick manifold block used for attachment of inlet and outlet connections. The second was for the between the reaction channels. The third was for between the coolant channels. The fourth was for the final lower coolant channel which met with a cap at the bottom of the device that did not require any holes for reagent entry or exit.

We ran four gasket design programs with the laser, each a varied template with a mix of copper gasket patterns. The foil, shipped as flat sheets and not in roll form, was secured to the glass surface with kapton tape. We also moved the emission filter hose away from the gaskets. The laser then cut out the copper gaskets. We were able to produce four gaskets per each 6-inch square of copper.

Forceps were used for retrieving the copper from the laser. We exercised extreme caution when handling the copper foils. We needed to handle them as little as possible to prevent folding and wrinkling of the foil. We wore nitrile gloves to prevent oils from our hands contaminating the surfaces of the parts.

Acetone removed surface contaminants such as dirt and oil from the foil extrusion processes needed to manufacture the foil. Acetone was applied using optical plastic foam lens swabs used for cleaning optical equipment such as photo development machinery and cameras (Liberty Photo Products, San Clemente, CA).

Different gloves should be tried because we found that acetone goes through and degrades the nitrile material. This did not make a noticeable impact to our work though, but it could signal that some nitrile material could have contaminated the copper foil or on the aluminum parts prior to bonding.

We placed the copper gaskets between the aluminum laminae in the pressing block fixture with alignment pins to keep the foils and lamina in tight registration. Layer by layer, the laminated device came together.

The vacuum furnace press received the now assembled device and fixture package, and processed the device into a single part. After processing, we tested the part and found that the device still leaked.

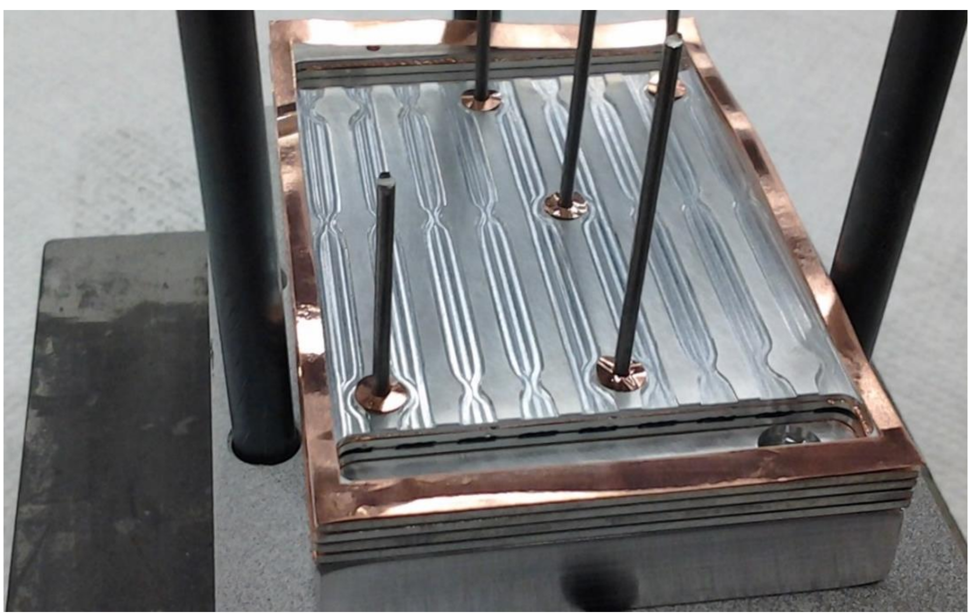

Figure 5-18 Copper foil interlayer variant placed between reactor laminae

We also noticed that the coil, with more copper mass, created much more eutectic alloy product than we initially anticipated. There were obvious solidified masses of eutectic alloy in the two wells of the unit and some dribbling on the outer perimeter of the unit. We would need to make an adjustment in the parameters to account for the added copper. We backed off on the amount of time and also minimized the amount of copper inside the device. We originally has copper fingers within the device to support the gaskets so that they were one part per layer. We minimized the amount of copper and only placed copper on critical bonding surfaces. This minimized the amount of eutectic product produced.

We also noticed that the module was not completely straight. Vertical alignment had been lost during the high-temperature bonding process. We previously noticed some bowing in the individual laminae, but thought that the heating process would soften the aluminum and allow them to bond under the load force of the press. Instead, the module emerged with a slight tilt. We needed to find a way to straighten the laminae prior to assembly in the vacuum furnace press bonding-fixture.

EUTECTIC BRAZING (ADDING AN ANNEALING PROCESS)

We noticed that after machining the features into the surfaces of the aluminum laminae, they exhibited a slight curvature. At first, we thought nothing of the curvature. We assumed the eutectic brazing process would correct this curvature since the device would be under some load and because our fixture used several alignment pins to keep the parts registered. We found that after bonding, our devices produced had a slight lean to them and the parts were still leaking. The misalignment of the device could open up holes between laminae, possibly enough to create leaks, and the movement could also misplace or tear the copper foil gaskets.

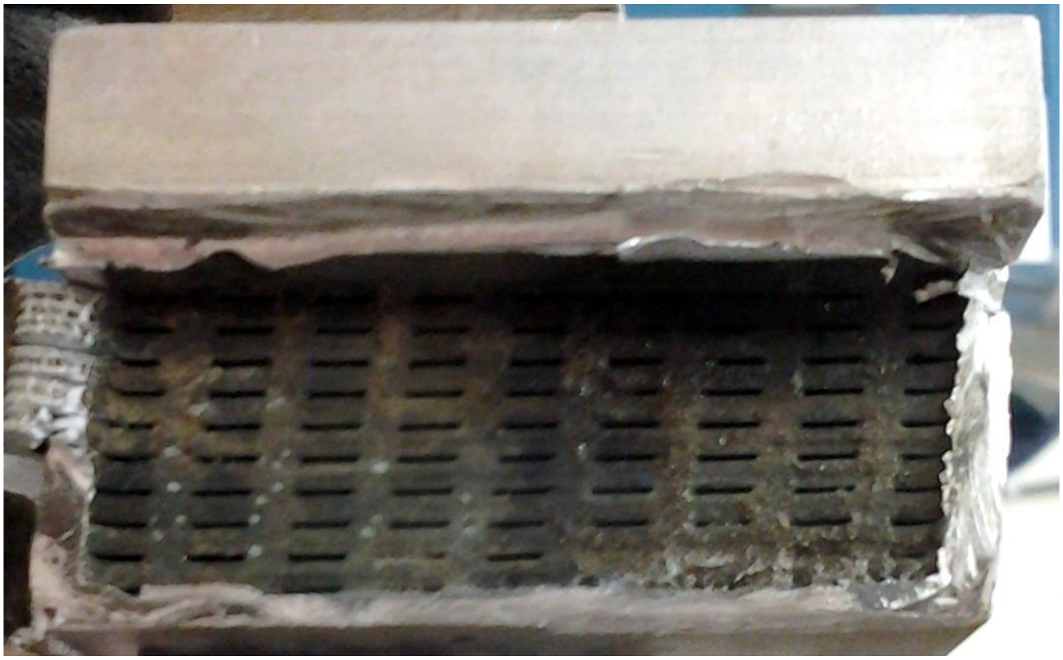

Figure 5-19 Evidence of deformation caused by machining stresses

The machining process introduces the curvature into the laminae as the milling process places stress on the surface of the part in the direction of the cut as the endmill climbs into the material. This forces the aluminum to split amongst the flutes of the end-mill. The stresses build up in the surface of the material and cause the warpage to occur. The curvature can be relaxed with a heat-treatment process to relax and anneal the metal.

First, we cleaned the aluminum laminae with acetone to remove any grease or machining coolant residually on the surface of the aluminum. We rinsed the laminae in deionized water. We used a sonicator to wash the laminae and utility air to dry the laminae.

We stacked the laminae (without the copper foil interlayers) in bonding order vertically within the bonding fixture that we would later use to bond the device. We used a hot-press (Carver inc., Wabash, IN) at 150C to relax and anneal the laminae. We applied approximately 80kg (175 lbs) of force. This process held the laminae in registration and in joining order for 3 hours. The block cooled in air after being removed from the hot press.

The stack of aluminum emerges as block that seems to be bonded. Friction caused by the thermal expansion and possibly the further evolvement of the aluminum oxide film on the surface held the stacked laminae tightly together. When held for longer hours (6 hours) the block becomes extremely difficult to pull apart. Even so, the block of aluminum pulls apart layer by layer with a crisp snapping sound.

Perhaps future work could explore the extent of this temporary bond. The straightened and annealed parts were then used for bonding. Unfortunately, leaks were still not eliminated.

EUTECTIC BRAZING (ADDED MECHANICAL POLISHING)

To improve the intimacy of the bonding surfaces, we thought it would be beneficial to polish the aluminum laminae. We first explored the use of electro polishing, but after witnessing the process in a laboratory, it became clear that electro polishing was difficult to apply to specific regions of the aluminum alloy part. We witnessed uneven erosion of the part. The chemicals needed to electro polish are also quite caustic [11].

We tried mechanical polishing using 3M automotive wet-dry sheets. We used various grits from 400 to 2000. We wet-sanded (or mechanically polished) by hand the aluminum alloy machined laminae in cold tap water. We followed the sanding by a wash of acetone, then deionized water, and then dried with plumbed air.

Figure 5-20 Polished aluminum alloy microscopy.

The surfaces for bonding were now smoother than before. We hoped this would contribute to a leak-free bond, but the problem of interior leakage still appeared in testing. We were still producing parts that had small, if not almost indiscernible, leaks. We would finally catch a break after an accident during the annealing process occurred.

EUTECTIC BRAZING (MADE A DESIGN CHANGE)

Through accident during a lamina annealing process, the machine place 20,000 lbs. of force upon the stack of laminae, heavily deforming the aluminum laminae. The force deformed the parts making them useless, but as we separated the parts, we were able to understand the load-transfer the device would undergo during the eutectic brazing process. This allowed us to understand why we were still having difficulty eliminating leaks from the interior channels of the aluminum laminae. The design prohibited the application of force in some critical bonding areas beneath the locations of the inlets and outlets.

The areas around the inlets and outlets were not backed underneath with supporting material. When the part was pressed under heat in the vacuum hot press, the laminae would not receive an even distribution of force. Areas that received less force were less likely to seal. When this was realized, the orifices were made smaller so as their openings did not overlap with the walls of the micro-channels located below in the adjacent laminae.

After this change, the inner-seal problem disappeared, and we gained our first of several successes in performing a eutectic braze seal of the laminated micro-fluidic device. We were able to repeat the previous processes and continued to produce positive results with our method.

WELD CHARACTERIZATION

The key to the development of the lamination method described in this paper involves eutectic brazing of the faces of the machined aluminum laminae with a copper foil. This lamination process eliminates the need for specialized fluxes, chemical surface treatments, masks, etc.

We started with two distinct materials: aluminum 6061 base material and copper foil. During the brazing process, we expected the concentration of copper to diffuse from the center of the joint outwards in to the aluminum, forming various eutectic alloys.

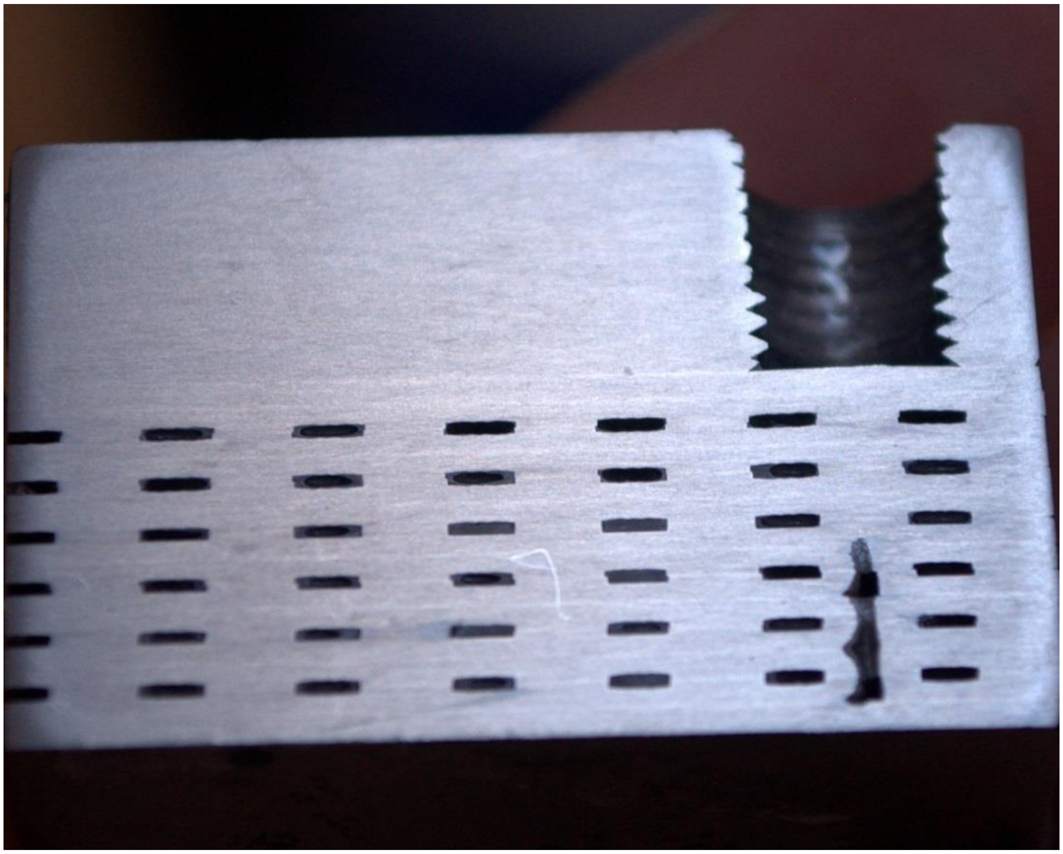

Figure 5-21 Cross-view of welds of laminated device

Using EDAX Energy Dispersive Spectrometry, we can characterize the elemental composition at specific points across the surface of a joint cross section. The detection of various levels of copper in the surface will allow us to understand the diffusion of copper, by the evolution of various eutectic alloys, into the base aluminum metal. This data allow us to understand the composition of the weld and compare it to the base aluminum 6061 alloy.

EDS detects x-ray emissions given off of the specimen when energized by the SEM. The model EDS System is an EDAX TSL with an Ametek 204B+ sensor. The bundled of the EDS identifies characteristic x-ray voltage pulses given off by each individual element. We used the EDS to measure energy levels associated with

Aluminum, Copper, Magnesium, and Silicon. We matched the energy levels to specific points across the weld and adjacent weld interaction areas. We wanted to know the characteristics of the weld such as the distance of element diffusion.

The images will show that the two aluminum alloy pieces can be considered as one. Aluminum alloy 6061 typically contains a range of copper between .15% to ,40% [5 (ASM)]. The eutectic brazing process produces a eutectic alloy with a slightly higher copper content than the aluminum 6061 alloy base material.

With this work, we learned about how difficult the process of lamination can be, not just in design and manufacturability, but in testing and verification. One of the difficult tasks for this project is analyzing the weld throughout the part. To analyze, the weld, the part must be cut apart. We used wire electric discharge machining (EDM) to slice through the part and prepare a specimen to analyze the eutectic welds. We then mechanically polished the specimens using the same method previously used to prepare the individual laminae for bonding.

Aluminum 6061 is a soft alloy, with large constituents of Magnesium and Silicon. Aluminum 6061 also contains small amounts of Copper, typically 0.04% to 0.1% by weight. The soft material has to be polished with a light hand and a controlled increase of grit. The following specimen was polished to 2000 grit.

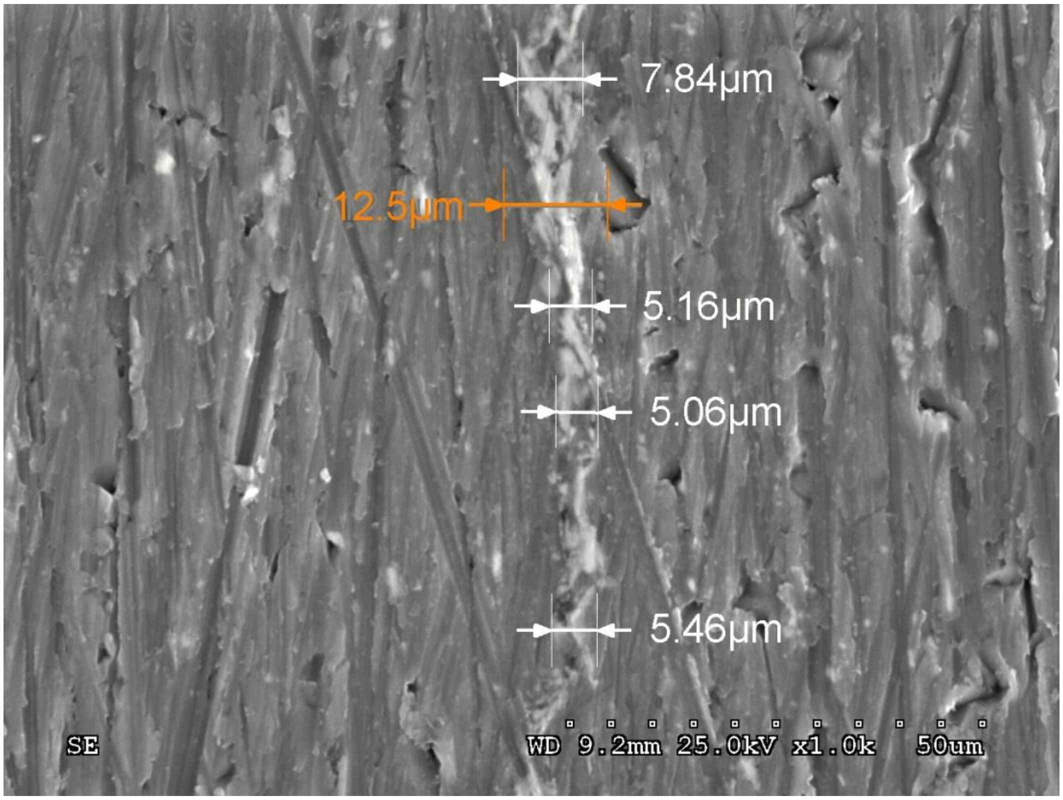

Figure 5-22 Measurements of the eutectic weld seam

Scanning electron microscopy (SEM) was used to capture micrographs of the weld between laminae. An example measurement of the original thickness of the copper interlayer foil, 12.7 micrometers, is displayed in orange. The exposed weld seam now appears several microns thinner than the starting copper interlayer material, leading one to assume some diffusion of the copper and aluminum has occurred. The higher reflectance of the copper illustrates that this 5.5 micrometer width contains higher concentrations of that element which will need to be quantified further.

The SEM we used was a thermal imaging Mitsubishi Model #S3000-N. The images are produced by the detection of electrically energized regions of the specimen within a vacuum chamber. The visible weld seam has a higher copper concentration in some parts, causing a lighter metal color to appear, as copper conducts electricity with less resistance compared to aluminum and aluminum oxide.

Through the use of EDAX Energy Dispersive Spectrometry (EDS), we show that indeed the elements have diffused. The specimen was measured for elemental analysis of the exposed polished surfaces approaching the weld seam.

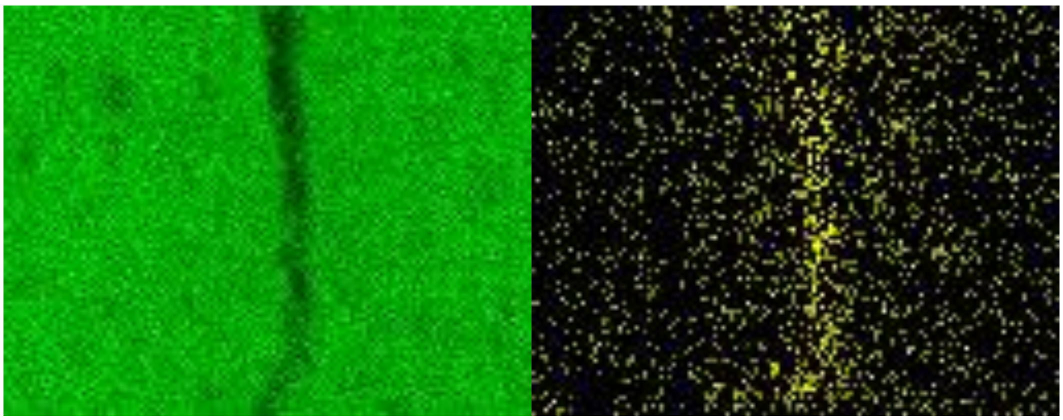

Figure 5-23 EDS surface detection of aluminum (left) copper

We notice that in the seam, aluminum, magnesium, and silicon have been detected. The original copper interlayer was 99.9% pure copper. We can see throughout the weld area that diffusion has occurred as the image shows lower concentration of copper and a high concentration of aluminum in its place. If no diffusion had occurred, there would be far less aluminum detected in the weld line. We will now look at measurement across the weld.

Five detection lines across the weld taken with the EDAX Energy Dispersive Spectrometry system produced profiles for further elemental analysis. The system trances lines drawn by a user in the software and reads them in increments of about 1.3 micrometers. The system measures the percent of each metal detected at the surface.

EDS uses X-ray detection to identify material composition from the surface of the specimen. The system then translates this information into information regarding surface element composition.

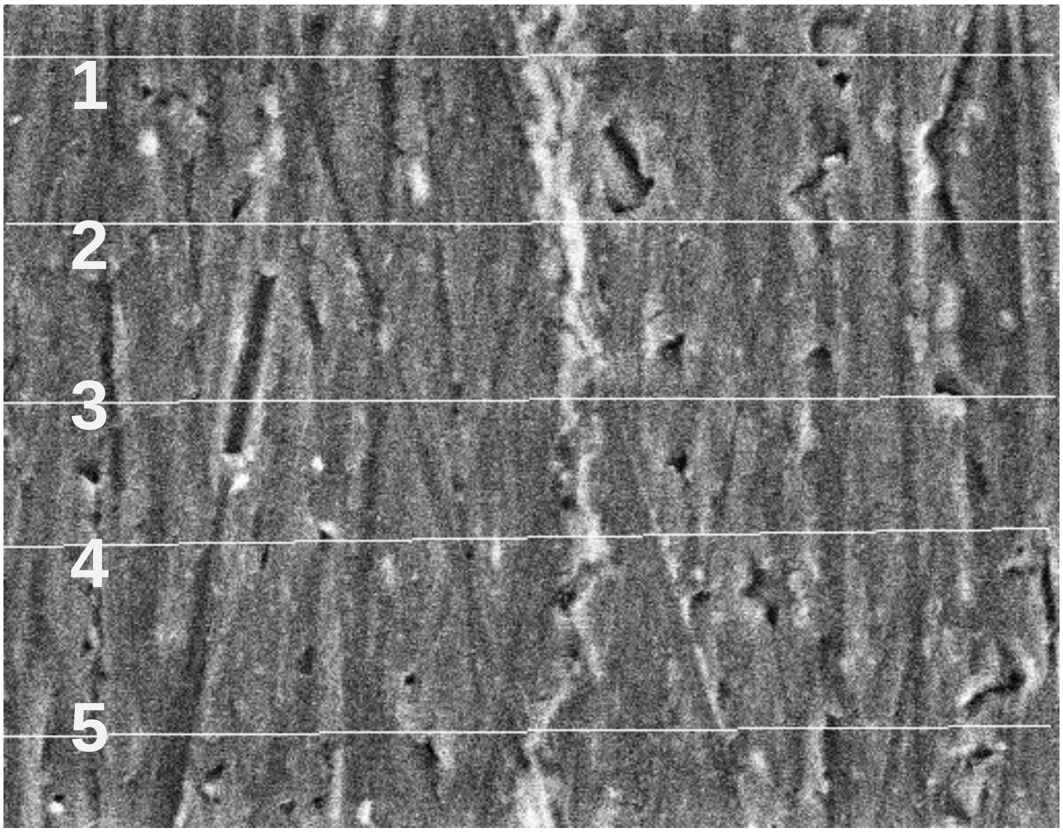

Figure 5-24 Lines of scanning for EDAX Energy Dispersive Spectrometry

The following charts shows the readings taken by the EDS system at each of the five white lines on Figure 5-24, from top to bottom. Measurements taken follow the user-drawn line from left to right. These traces produced the following information.

In Figures 5-25 & 5-26, line 1, the detection of copper rises sharply between 272.56 micrometers and decline until 327.6 micrometers of distance. This diffused weld is approximately 55.04 micrometers wide.

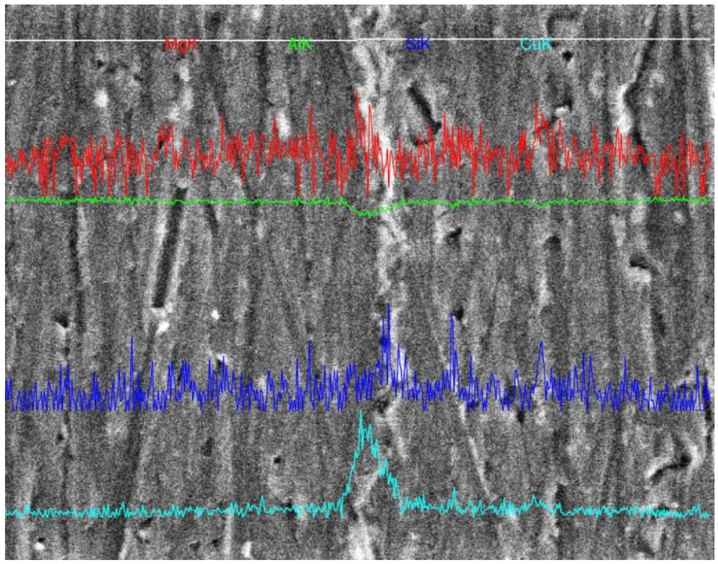

Figure 5-25 Line 1 - EDS element detection of line 1 with SEM overlay

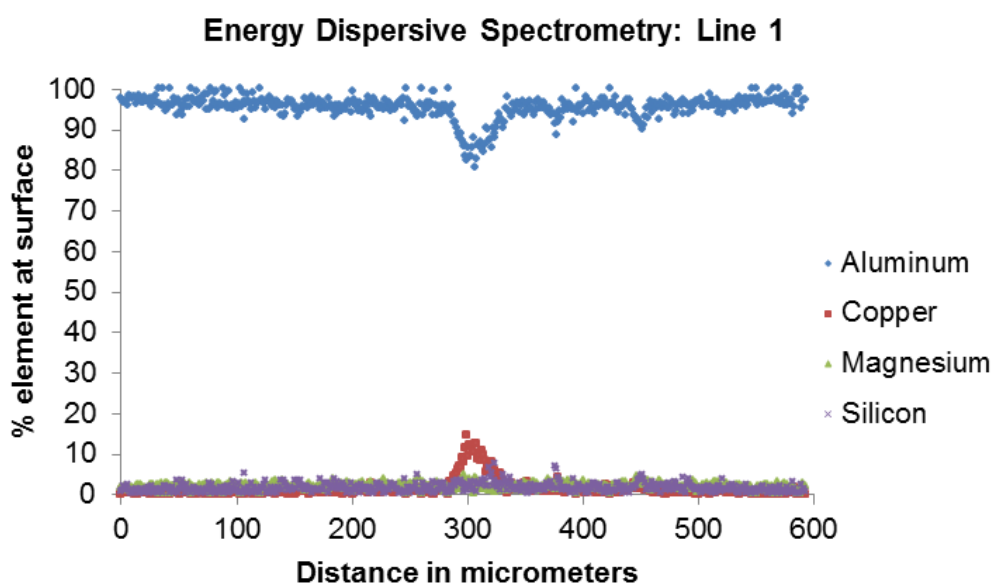

Figure 5-26 Line 1 - EDS element detection of line 1 percentage (%)

In Figures 5-27 & 5-38, EDS detection along line 2 shows that concentrations of copper rise sharply from 283.04 micrometers and decline until 342.45 micrometers of distance. This diffused weld measures approximately 59.41 micrometers wide.

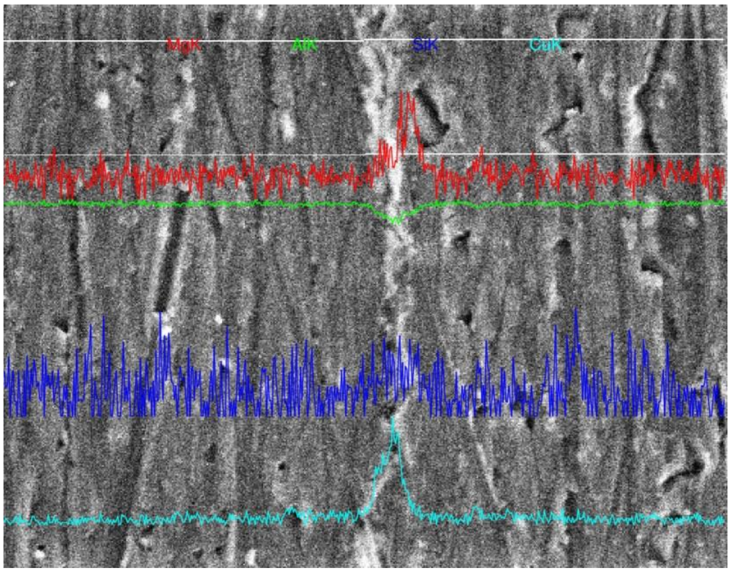

Figure 5-27 Line 2 - EDS element detection of line 2 with SEM overlay

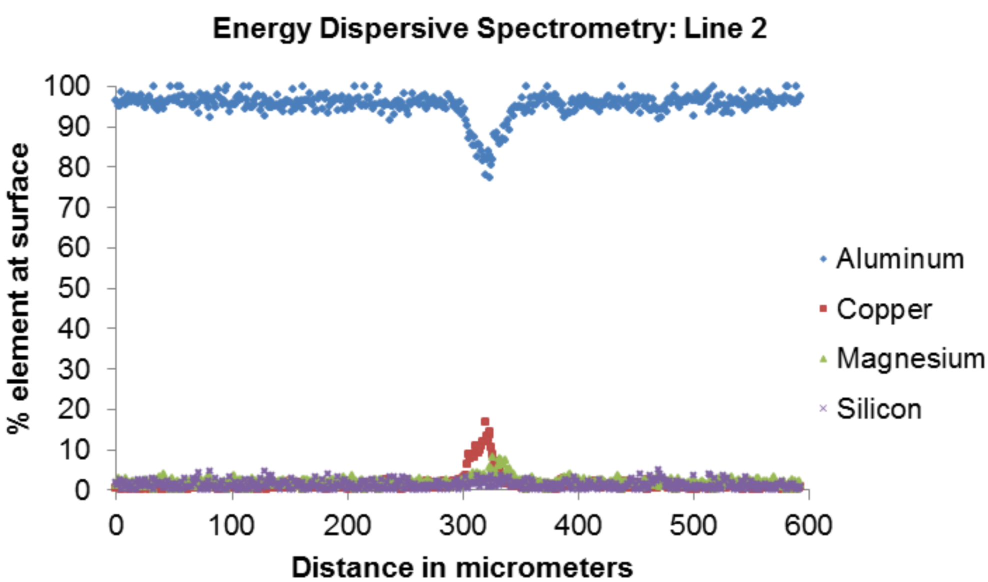

Figure 5-28 Line 2 - EDS element detection of line 2 percentage (%)

In Figures 5-29 & 5-30, EDS detection along line 3 shows that concentrations of copper rise sharply from 299.35 micrometers and decline until 338.96 micrometers of distance. This diffused weld measures approximately 39.61 micrometers wide at this location.

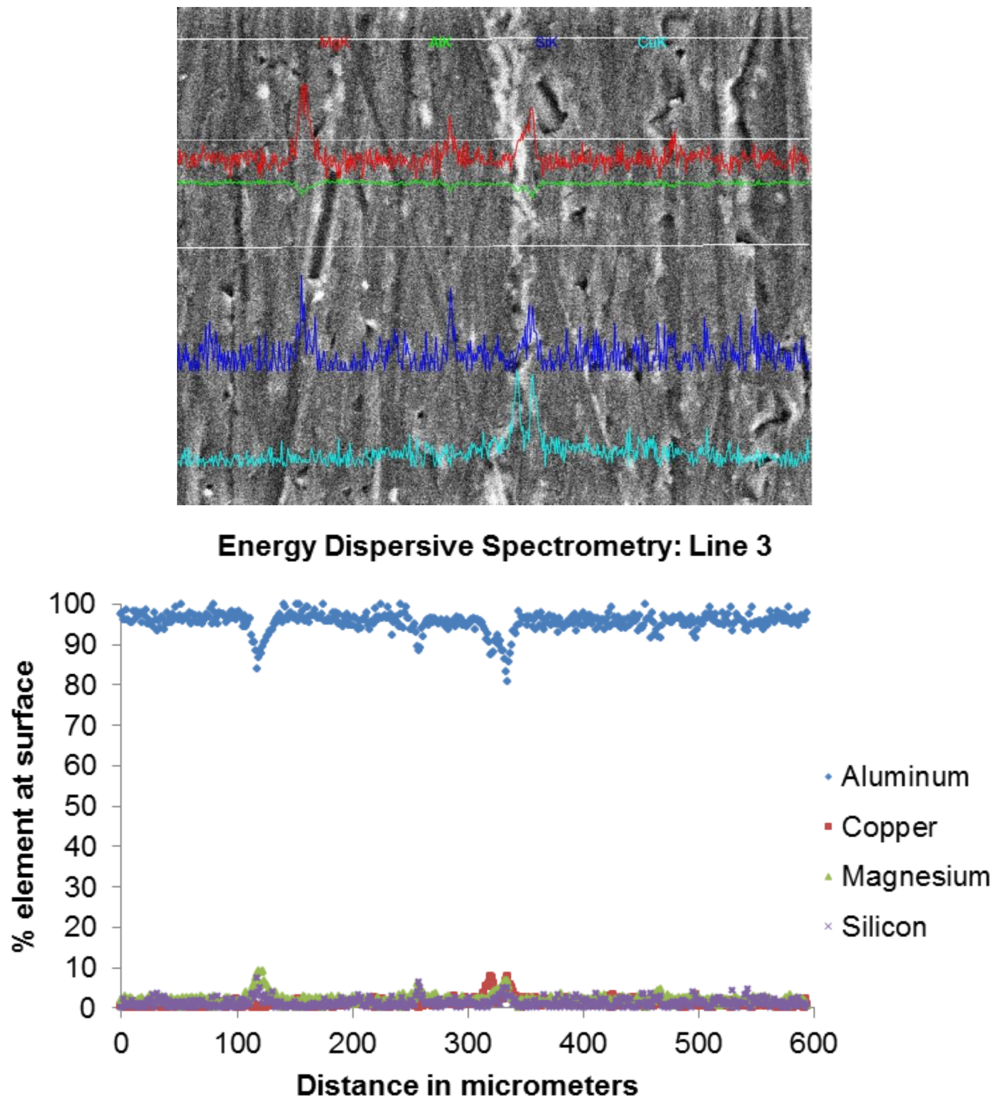

Figure 5-29 Line 3 - EDS element detection of line 3 with SEM overlay

Figure 5-30 Line 3 - EDS element detection of line 3 percentage (%)

In Figures 5-31 and 5-32, EDS detects along line 4 that the copper is concentrated between 305.23 micrometers and declines until 344.94 micrometers of distance. This diffused weld measures 39.71 micrometers wide.

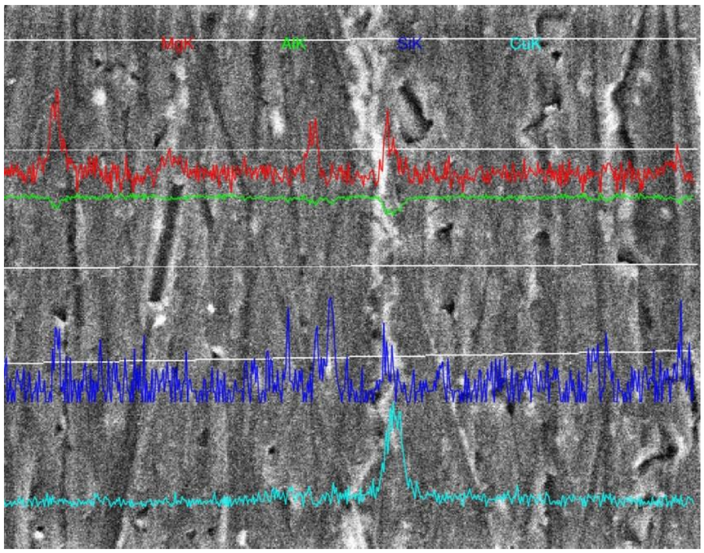

Figure 5-31 Line 4 - EDS element detection of line 4 with SEM overlay

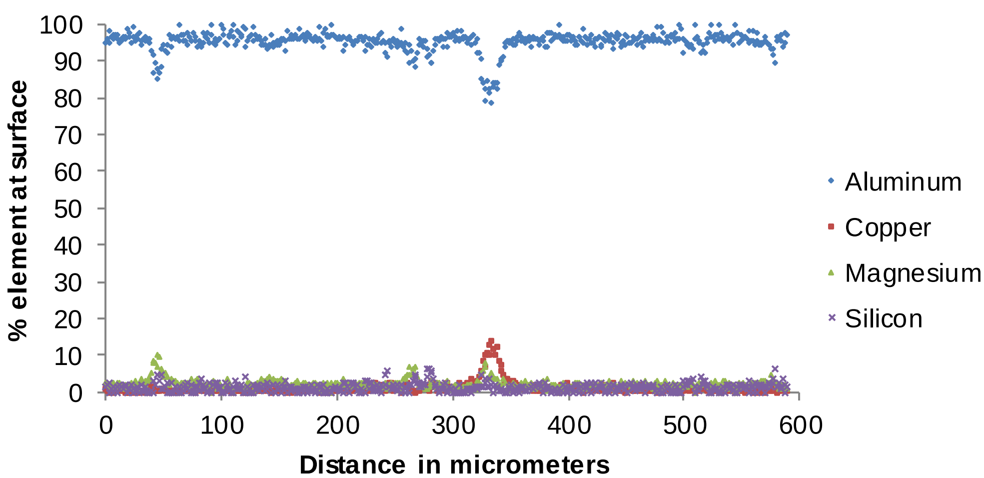

Figure 5-32 Line 4 - EDS element detection of line 4 percentage (%)

Along line 5, in Figures 5-33 and 5-34, EDS detects copper concentrated between 281.89 micrometers and 308.69 micrometers of width. This diffused weld measures approximately 26.8 micrometers wide.

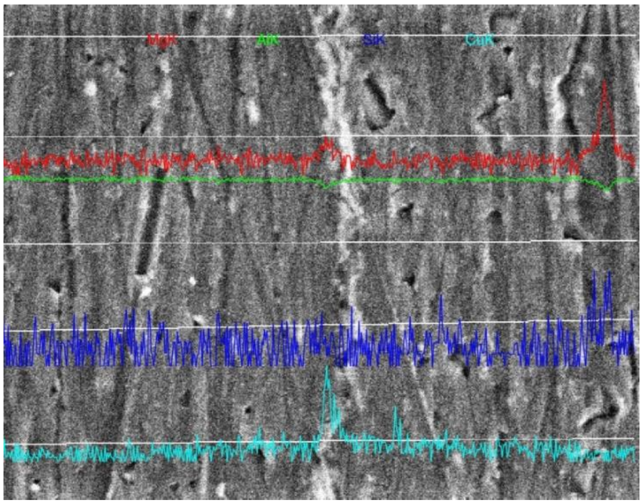

Figure 5-33 Line 5 - EDS element detection of line 5 with SEM overlay

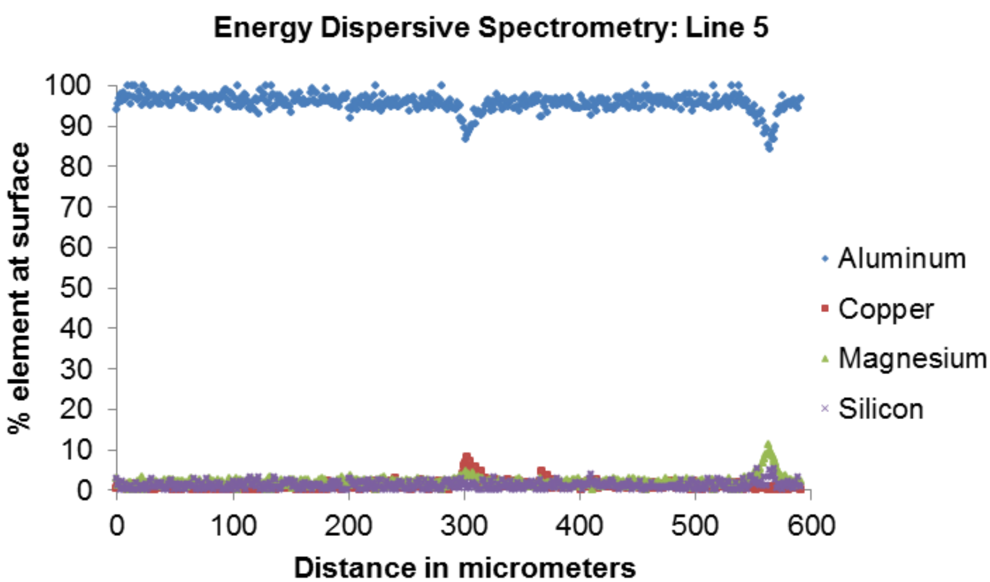

Figure 5-34 Line 5 - EDS element detection of line 5 percentage (%)

With this information, we can claim that diffusion among aluminum and copper has occurred. At the weld seam, nearly 80% of the detected material is aluminum compared with less than 20% copper. Both materials have diffused into eutectic alloys near the weld line. The weld line itself, measuring from the abrupt rises in copper and dip in aluminum content, has spread from the 12.7 micrometers of the copper foil to approximately 44.12 microns in width. That is 3.47 times the original width of the eutectic copper interlayer foil. This is evidence of eutectic diffusion.

Chapter 6

Final Process

The literature review revealed several contemporary joining methods for metals. Several processes were identified as possibly working to join aluminum laminae. We looked at direct diffusion, adhesive, brazing, soldering. After experiments, we found that eutectic brazing created a bond that was strong enough to hold up to the system testing under temperature and pressure. We did not try soldering or brazing with flux. Upon discovering that eutectic brazing produced a decent bond that eliminated a majority of the leaks, work focused to completely eliminate leaks.

After several process changes and a design correction, we produced several microfluid devices that were leak-tight. These devices were used in synthetic gas to liquid fuel experiments.

We will now discuss the series of processes, consisting of machining, annealing, cleaning, polishing, stacking, and processing, that produce a positively sealed microfluidic reactor device.

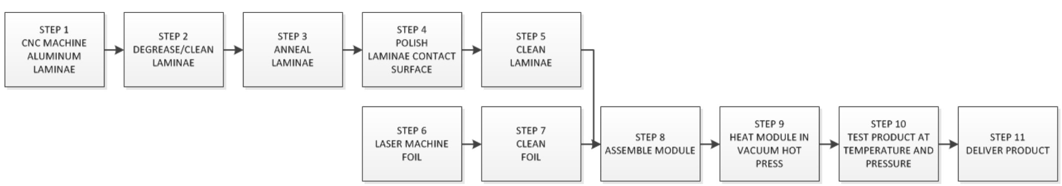

Figure 6-1 Flowchart: process steps

There are 11 main steps to the eutectic brazing procedure. Before the process begins, various supplies and tools need procuring to prevent process starvation. Supplies

An inventory should be made of parts & supplies necessary for the manufacturing project. The following supplies produce the machined aluminum laminae: aluminum alloy 6061 raw cut stock, endmills, and coolant for the CNC endmill machining center.

Endmill selection should match the larger features of the work as closely as possible, narrowing as necessary to minimize the tool changes and tool path length. This will help curb endmill wear and cycle time.

Producing copper foil interlayers for the copper-aluminum eutectic braze requires the following supplies: copper foil stock, polyimide tape to hold down the copper foil in the laser-machining center, and a fixture or substrate to attach the copper foil.

Cleaning the various aluminum parts and foil gaskets required the use of several chemicals, supplies, and machines. The endmill machining center floods coolant over the working part and the endmill to extend the life of the tool. The aluminum laminae we used arrived with an oily machining coating from manufacturing and now received this coolant flood coating.

These types of coatings can off-gas or carbonize when exposed to vacuum or heat, respectively. We used a commercial degreaser, an automotive aluminum wheel cleaner (Eagle One Aluminum Wheel Cleaner), to clean the laminae in preparation for annealing, and washed the laminae in deionized water to remove any dust and detergent.

After removing dirt and grease, we used a simple registration fixture and the Carver hot press to relax and anneal the aluminum alloy laminae. The same fixture would be used with the vacuum furnace press. The fixture has alignment pins and ensures the aluminum parts do not slide around during the straightening and annealing process.

The fixture transfers heat and load evenly among the aluminum laminae. We reused the same fixture used for bonding the parts later in the vacuum hot press. The fixture comprised of two thick carbon steel blocks with alignment holes for six carbon steel alignment pins. The pins attached to the lower carbon steel block with a circumferential friction fit and featured a taper at the ends that allowed for the opposite block to slide away to release the stacked laminae before and after annealing. After annealing, we mechanically polished the straightened aluminum parts by hand.

Numerous levels of sanding paper deliver the most controlled approach of mechanically polishing, ensuring a consistently declining roughness to the finishing grit roughness. The grit in higher-number-grit sanding sheets will tend to be powder-like. In our experience, these sheets wore out faster than the rougher, lower-grit sanding sheets. It is best to have extra supplies of the higher-grit sanding sheets. We found that 3M automotive paint preparation wet and dry sanding paper featured consistent grain sizes and noticeable durability compared to other brands we tried.

After sanding the parts, acetone, followed by deionized water helps remove any organics possibly introduced during the mechanically polishing, and washes away contaminants and dust. We used filtered utility air to dry the parts.

Gloves and protective masks should be used at all times, and the cleaning should take place in a well-ventilated area. Avoid the use of paper towels for drying parts and instead use compressed or utility air (with an oil trap). Paper towels will introduce organic fibers onto the laminae creating an opportunity for pyrolysis, which could lead to poor braze seals. Avoid organic contamination from fibers and body oils.

The vacuum hot press requires servicing of a gasket on the underside of the chamber door. Spread silicone vacuum grease along the gasket to ensure a good seal and to extend the life of the gasket. The vacuum hot press also consumes a fair amount of silicone oil in one of the vacuum pumps.

The vacuum pump, called a diffusion pump, operates during the second stage of the vacuum process after the conventional vacuum pump (a displacement pump) reaches its lowest level of vacuum. The diffusion pump works by trapping gas within the silicon oil and passes it through as an exhaust, acting as a sort of barrier between the vacuum environment and the outer atmosphere. This produces extremely low levels of vacuum, approaching 10^-8 mbar. These low levels of vacuum heavily reduce the rate of oxidation in the chamber. This makes it possible to perform high-heat materials processing without the worry of oxidation.

The testing phase requires the use of argon, a thermometer, silicone oil, and a heating plate. We will now discuss the various process steps in detail.

STEP 1 CNC MACHINE ALUMINUM LAMINAE

Numerically controlled endmill machining center transforms blank aluminum coupons into machined laminae. The device consists of 5 separate types of parts. We will refer to these parts as A through E. Two part designs, C & D, are repeatedly produced to increase the number of parallel reactor and serpentine coolant laminae layers.

Part A, the upper manifold part contains holes for inlets and outlets, alignment pin holes, and threads for the fittings. There are two separate systems – reagent and coolant. Each system has its own threaded inlet and outlet.

Part B sits below Part A. The design for Part B specifies a one-side machined part. The top has no features and the bottom has one serpentine coolant channel. Part B has holes for inlet and outlet flow and holes for alignment.

Part C is a two-side-machined with a parallel group of reactor channels on one side and a serpentine coolant channel on the reverse. The part has a large trough-like hole at the end of each side to allow for reagents and products for all laminae layers to flow.

Part D is identical in number of channels and holes, but with all machined features mirrored about its longitudinal axis to match up with Part C.

Part E sits below all of the parts. It is a longitudinal mirror of Part A, but with all through holes not needed for alignment deleted.

The parts are produced by translating drawings designed primarily with computer aided drafting (CAD) software package which are then translated into machine code through a post processes on the back end of a computer aided machining (CAM) software package. In our case, GibbsCAM was used to interpret the Rhino CAD models into a range of endmill tool paths and programs.

These toolpaths control the NC endmill machining center to produce the parts.

The NC endmill machining center produces the parts. We used a 3-axis NC machining center. Each machined laminae required flipping by hand that could also be automated by a 5-axis NC machining center. The parts took about 45 minutes combined to machine both sides.

STEP 2 DEGREASE & CLEAN LAMINAE

We used ultrasonic cleaning when degreasing the aluminum parts prior to sanding. The parts typically spend about 10 seconds in the ultrasonic cleaner before moving to another ultrasonic cleaning bath of deionized water for another 10 seconds. We then removed the parts from the deionized water bath and dried them using forced air. We then took the parts to the next station for mechanical polishing.

STEP 3 ANNEAL LAMINAE

Using the Carver hot press, and using the bonding registration fixture, we loaded the assembled laminae in order to heat them for annealing. The parts were heated at 150°C and held for 3 hours. It is important not to exceed 6 hours of holding time as the aluminum oxide grows and makes it difficult separate the laminae.

The fixture for holding the laminae during bonding was constructed out of carbon steel. The fixtures consists of two 1.5 inch thick blocks of steel with six alignment pins.

The pins are tapered to allow one of the block to be removed. A friction fit of the opposite end of the alignment pins provides rigidity to the lower part of the steel fixture.

Steel was chosen due to machinability, high-temperature use, low cost, and rigidity. The steel fixture did not deform or mark throughout its use after carrying out many heating and cooling cycles.

After annealing the laminae, we removed them from the fixture and prepared them for mechanical polishing.

STEP 4 POLISH LAMINAE CONTACT SURFACE

Using the 3M automotive wet-dry sanding paper, we mechanically polished the aluminum 6061 laminae bonding faces under cool running water. We controlled the grit size from 600 up to 2000 grit. Polishing improves the contact surface by removing burrs and scratches, as well as extra oxidation film built up during the annealing process.

STEP 5 CLEAN LAMINAE

We cleaned the laminae in a sonication of acetone to dissolve surface organics and rinsed the laminae in deionized water. We made sure to keep each laminae in perfect order and orientation. Once cleaned, we prepared the laminae to receive the laser-cut copper interlayer foil.

STEP 6 LASER MACHINE FOIL

We machined the copper interlayer foil using the UV laser machining center. The machining center works similarly to an NC machining center in that it requires CAD and CAM processes to create workable machining programs. We built three separate machining programs to produce the necessary copper foil gaskets for our microfluidic device.

Copper foil as an interlayer between aluminum alloy 6061 allowed the brazing together of aluminum alloy 6061 laminae. While copper has a melting point of 1080 degrees Celsius, the joining of aluminum and copper together requires heating to a lower temperature in which a eutectic process occurs. The eutectic point, or lower melting point between binary alloys of copper and aluminum, exists at 550°C.

STEP 7 CLEAN FOIL

The laser machining copper foil received a wipe down with acetone with optical swabs. The aacetone removed surface contaminants such as dust and oil from the foil extrusion used in the manufacturing plant. We used optical plastic foam lens swabs to apply the acetone. We chose swabs commonly used for cleaning optical equipment such as photo development machinery and cameras (Liberty Photo Products, San Clemente, CA). They are made of plastic foam that leaves no lint.

STEP 8 ASSEMBLE MODULE

A coating of boron nitride (ZYP Coatings, Boron Nitride (BN) Aerosol Lubricoat® spray can) acts as a release agent between the steel and the aluminum. The coating prevents the steal and the aluminum from bonding (chemically or by surface friction) and prevents any drips of the eutectic material from bonding to the steel fixture as well. This should be sprayed on the fixture (not the parts to be bonded) before the laminae and the copper interlayer foil gaskets are assembled.

The process for assembly requires the use of a registration fixture such as that we used for the vacuum furnace bonding and the annealing process. The parts need to be held in registration throughout bonding.

Assembly takes place by stacking the parts in order of bonding. We recommend orienting the device so that the inlet-outlet parts are on the topside rather than below. This prevents eutectic material from seeping into and blocking the holes.

STEP 9 HEAT MODULE IN VACUUM HOT PRESS

After the laminae parts are combined in the fixture, the entire unit enters the vacuum hot press. The vacuum hot press will vacuum down the unit to 10^-4 Torr. The heating elements then activate to bring the chamber to 560°C at a rate of 30°C per minute. The chamber will hold the device at temperature for 30 minutes. We allow the chamber to cool on its own (the walls of the chamber are water cooled). After about an hour, the chamber will reach a temperature safe for people to access the contents. We then device and let it cool before carrying it to our test bed.

STEP 10 TEST DEVICE AT TEMPERATURE AND PRESSURE

At the testing station, the completed device connects to the argon tank with stainless steel tubing and fittings at one of the inlets. A Stainless steel plug fits into the corresponding outlet. The argon tank pressurizes the unit to 160psi. The unit is placed in a bath of water and pressurized to see if any obvious leaks occur. The unit is then removed from the water bath, dried, and then enters the silicone oil bath.

The silicone oil and module are allowed to steep for 3 hours. An observation is made to decide if the device is leaking or not.

STEP 11 DELIVER PRODUCT

If the test is successful, we remove the module from the heated silicone oil bath, making sure to allow it to drip residual silicone oil back into the silicone oil bath. We then wash the module with soap and water, making sure to keep the inlet/oulet holes plugged to prevent water or oil from seeping inside. The module is then ready to be used for fuel experiments or further study.

SUMMARY

We found eutectic brazing to be a suitable joining method for the aluminum 6061 laminae. The latter part of the work focused on eliminating all leaks and producing a usable microfluidic devices made from several layers of aluminum laminae. The device was produced with a vacuum hot press thus prohibiting oxidation and not needing flux. The copper in the interlayer foil diffused into the aluminum oxide layer, breaking it up, and allowing the evolution of a copper-aluminum eutectic alloy to form. The copper-aluminum eutectic alloy provides a seal that allows the laminated aluminum 6061 device to operate at temperature and pressure for many hours without failure.

Chapter 7

Conclusion

We identified different methods that would allow us to join the laminae. The restrictive geometry of the laminae faces made difficult the use of various contemporary joining methods, such as welding, soldering, adhesives. We found that it was possible to braze the parts with a eutectic alloy using commercially available copper foil as an interlayer between the laminae of aluminum 6061. The copper and aluminum 6061 materials produced a eutectic alloy bond, sealing the part hermetically, and allowing the use of the device with high-temperature and high-pressure.

We were able to produce the laminated microfluidic reactor device using contemporary machining techniques. Conventional NC endmill machining formed the aluminum 6061 parts. An Nd:YAG Laser machining produced the copper foil interlayers. A vacuum hot press allowed the bonding of the laminae in avoidance of oxidation. These machines can be acquired for a sum less than $1M.

Our specific objective for this work was to produce a method suitable for producing an aluminum alloy 6061 microreactor device. The device needed to seal two separate channel systems in close proximity to each other to facilitate the hydrogenation of synthetic gas in the presence of a catalyst.

We met several challenges with diffusion welding and adhesive, but found that eutectic brazing brought us closer to success. Working with eutectic brazing, we adjusted the interlayer thickness, adjusting the mass of copper in the device, and adjusted the process to facilitate the joining of the laminae. We adjusted the shape of the laminae and the surface morphology with annealing and mechanical polishing. We eventually came to a conclusive method that produced a device with an hermetic seal.

We show evidence of this seal through the leakage testing carried out in our laboratory, micrographs of the copper-aluminum eutectic weld, and pointing to related work in the literature featuring a eutectic brazing process.

Our devices were used for testing hydrogenation of synthetic gas in the presence of an embedded catalyst. No reports of failure (leaking) have been made at the time of writing this paper.

The process only requires two steps that use heat. The first heats the laminae to relax the aluminum 6061 parts after being machined. Holding these parts anneals the parts and allows them to be flat. The second heating process is needed for bonding in the vacuum hot press. The vacuum chamber is heated with graphite electrodes producing radiant heat. The entire part is exposed to the heating process, with heat conducted through by the copper interlayer and aluminum laminae.

Dangerous acids and atmospheres are avoided with this process. Acetone and aluminum degreaser were used to remove machining coolant residue and dust from the mechanical polishing stage. No flux or detergents were needed. No clean-room processes are required with the final process.

Materials removed during processing, aluminum burrs and copper foil remnants, may be materials of value to recyclers.

Several approaches and observations to the problem of joining multiple layers of alloy 6061 together led to the use of eutectic brazing. Aluminum-copper eutectic brazing seals the micro-fluid reactor device between each laminated layer of reaction and coolant channels. Heating the aluminum laminae sandwiched with copper foil interlayers produces eutectic alloys with lower melting temperatures than the two separate parent materials, aluminum and copper. This process allows the machined features on the surfaces of the aluminum laminae to retain their shape as only the surfaces with direct contact between copper and aluminum react to produce eutectic alloys.

The final product, a method for bonding aluminum 6061 laminae with machined micro-channel features using eutectic brazing, can be used in further research and by commercial entities. Those looking to produce microfluidic devices should study eutectic brazing to see if it is suitable for their materials and uses.

Through this work, we have accomplished the assembly of a method for fabricating microfluidic reactor devices from aluminum 6061 laminae without the use of flux, cleanroom, or caustic chemical need. We were able to show that the diffusion of aluminum and copper occurred and that the devices built with this joining method were usable for laboratory experiments for synthetic gas hydrogenation.

If the project were funded further, we might look into figuring out ways to optimize material use and lower cost of producing the components. We would look into producing the aluminum laminae through other means such as photolithography or some other faster means. We would also look into figuring out if we could make a denser reactor with more channels to produce a higher level of throughput. We could also look at making the microreactor modules larger, scaled-up to produce more throughput.

Chapter 8

Future research

Future research needs to focus on the reduction of cost and processing time required for microfluidic devices. Future work could identify ways to lower manufacturing and material costs. Future work should investigate the capital investment requirements of mass-manufacturing microfluidic eutectic diffusion brazed devices [18] [20].

For example, future work needs to evaluate the optimum amount of copper needed to produce a strong eutectic braze joint to reduce material costs. Some ratio of the copper thickness to aluminum surface area may provide the optimal amount of material to produce a complete eutectic bond, further eliminating the weld seam and avoiding any erosion of machined surface features.

In addition, processing costs and processing time needs to be lowered considerably. Methods, such as etching and semiconductor methods need to be evaluated. Limitations of our arrived method are due to the use of a vacuum furnace. In an environment of mass production, a vacuum furnace may result in a bottleneck due to the long amount of time needed to remove the atmosphere from the heating chamber.

Another pass at the use of metal-ceramic adhesive should happen. The product likely works in many high-heat applications. If applied correctly, it may provide an excellent seal. This should be explored. Other rations with less liquid constituent may set up with better results than we had when we tried to apply the product to our aluminum laminae. An alternative application device, such as a stamp or a roller, or perhaps a silk-screening device such as that used for marking aluminum cans, could apply a thin consistent layer of the adhesive to the laminae.

Photolithography etching should be considered due to its high-output capability. Further cost analysis and design adaptation could adopt this method of production. This has the potential to quickly multiply production rates compared to conventional machining.

Also, the temporary aluminum oxide bonding we produced after 6 hours of our annealing process may be useful for some application outside of the scope of this project. The oxidation made the laminae extremely difficult to separate.

In space and low-gravity environments, such as The Moon or Mars, a useful role in manufacturing will be played by microfluidic devices. Microreactors facilitate reactions and mixing regardless of planetary gravity because of the microfluidic behavior or fluids in dimensionally constrained spaces such as microchannel. Microreactors help conserve input materials by allowing operators to produce exact quantities of products as needed. The products are of higher purity.

Microfluidic devices could find applications in various subsystems of extraterrestrial mission equipment. Systems for waste processing, energy production, fuel conversion, food production, food processing, will benefit from the use of microfluidic devices. These processes could pull from different feedstock and have them mixed and reacted within microfluidic devices, produced on-demand.

The devices, pumps, and containers required for microfluidic processing could also save weight by avoiding the need for large vats, large heating equipment, mixing devices, straining devices, and the respective cleaning equipment needed to maintain this equipment. Less weight and smaller physical size make microfluidic device attractive due to the lower payload launching cost.

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Biographical Information

Paul Wilson was born in Phoenix and has lived in Chicago, Albuquerque, and Fort Worth. Paul began studying Industrial Engineering in 2006. During the summer between sophomore and junior Industrial Engineering studies, Paul became involved with research and consultancy projects related to biodiesel, DFW International Airport operational safety, American Airlines Cargo priority parcel tracking.

Paul is an integral part of CREST (Center for Renewable Energy Science and Technology) research success which has gained millions of $ of research funding. His important role has been to fabricate prototypes and perform economic analysis of various processes, which are part of the proposals and reports to the funding agency. Coal-to-liquid and gas-to-liquid research fill the majority of his work. He is responsible for supervising two labs, which include laser machining, CNC machining, 3D microscope, hot press, and vacuum hot press. Paul supervises between two and five students every semester.

Paul's current research work is on building microfluidic devices in glass for biomedical assays, brain neuron drug interactions, and alternative fuels.

Paul joined the fast-track BS to PhD program in 2010 and defended his PhD dissertation in August 2013.

Paul's engineering interests are in manufacturing aspects of the space transportation, digital communications, and solar energy. Paul's hobbies include Kajukembo, cycling, sailing, and playing golf.