MICRO CHEMICAL SYSTEM DEVELOPMENT PROGRESS AT THE PACIFIC NORTHWEST NATIONAL LABORATORY

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Introduction

Microsystem investigations at the U.S. Department of Energy's Pacific Northwest National Laboratory (PNNL) are focused on the development of compact systems for chemical conversion/separation and energy conversion. Potential near-term versions of these systems include hybrids of conventional technology and microtechnology, adopting components that exploit the enhanced heat and mass transport available in engineered microchannels. Advanced microcomponents and systems at various stages of development include microchannel heat exchanges, reactors, gas absorbers, liquid-liquid extractors, heat pumps, and microactuators for pumps, valves and compressors. If successfully developed, the authors believe that microtechnology-based thermal and chemical components and systems may offer compact, distributed hardware solutions to energy and chemical conversion problems that previously have been addressed through the use of large-scale, central production facilities.

Research Approach

The microsystems research objective at PNNL has been to investigate the potential of microsystems technology for energy conversion and chemical processing. The overall approach for the last five years has been:

1) To develop micro thermal and chemical components performing a variety of unit operations, and

2) To develop system architectures which incorporate multiple microcomponents, collectively providing large-scale processing capability

Enhanced Heat and Mass Transport in Microchannel Arrays

The use of engineered microstructures offers the potential to reduce conventional resistances to heat and mass transport. For example, in the late 1980's researchers at Germany's Kernforschungszentrum Karlsruhe (KFK) demonstrated a compact, 1 cm^3 crossflow heat exchanger, using stacked microchannel arrays, that provided for nearly 20 kW_t of total heat transfer between the two working fluids.

More generally, heat transfer from solid walls to a fluid is limited by the laminar sublayer that forms within the fluid against the solid surface. Within a microchannel, the thickness of this sublayer is structurally constrained to less than the width of the fluid channel. An array of parallel flow paths, such as that depicted in Figure 1, therefore requires short fluid resistance times and can be expected to provide high overall heat transfer within a compact hardware volume.[1]

Perhaps as importantly, it may be generally possible to avoid high pressure drops (translating to high pumping power costs) within engineered microchannel arrays. This result can be obtained by engineering multiple, parallel microchannels, of high aspect ratiio, with short lengths. Confirming this, researchers at PNNL have demonstrated improved microchannel heat exchangers in a planar, sheet architecture, that

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[1] For laminar flow in microchannels, the time scale and the channel length necessary to achieve substantially complete heat transport are each proportional to $d^2/'\Upsilon$ and $Vd^2/'\Upsilon$, respectively, where d is the width of the channel, $'\Upsilon$ is the termal diffusivity of the fluid, and V is the mean velocity of the fluid.

exhibit relatively high heat fluxes (100 watts/cm^2 perpendicular to the sheet) and convective heat transfer coefficients (10,000 to 35,000 wats/cm^2-°C, or about one order of magnitude higher than typically seen in conventional heat exchanger hardware), with low pressure drops (2 to 3 psi). Typical microchannel heat exchangers are depicted in Figures 2 and 3. Results over the past four years have been obtained through utilization of a high-flux test loop for microchannel heat exchangers, and a number of geometries and channel/fin configurations are under investigation.

PNNL researchers are similarly developing microcomponents featuring enhanced mass transport. In 1995, as an initial, proof-of-principle experiment, enhanced mass transport within a ***microchannel gas absorber*** was investigated. Researchers demonstrated ammonia (g) absorption into water (1) with a 10 cm^2 test article at higher gas absorption rates than typically obtained with conventional technology.

Substantial heat was generated (approximately 30 watts/cm2), leading to the confirmation that an extremely compact, high throughput, microchannel gas absorber would require heat removal such as can be provided by the integration of a microchannel heat exchanger as part of the device. Figure 4 depicts the device; as is noted the liquid and gas phases are brought in contact through a microporous contactor plate, with surface tension keeping the liquid from leaking into the gas channel.

Likewise, researchers at PNNL are currently developing a compact separations system, for liquid-liquid separations, employing a ***microchannel solvent extraction contactor.***
The principle functions of this component are to 1) create a high contact surface area between the feed and the solvent streams, and 2) enable rapid mass diffusion by structurally constraining each streams' thickness. The constituent of interest, the solvent, would be quickly transferred to the solvent from the feed stream. While traditional solvent extraction contactor units require the introduction of mechanical work (i.e., partial mixing) to facilitate mass transport, the microchannel solvent extraction contactor (depicted in Figure 5) allows intimate contact between two thin films. Advantages include minimal dispersion of the solvent within the feed stream plus high mass transference rates per volume of hardware.

In general, the heat and mass transport experiments are confirming the ability to reduce transport resistances while not drastically increasing pumping power costs in an engineered microchannel. These results, are, in fact, consistent with conventional transport theory, and suggest the potential for compact, high capacity systems as solutions for thermal and chemical problems. The ongoing work includes the integration of microchannel heat exchangers with other devices, which will be discussed below.

Chemical Reactors

Researchers at PNNL have investigated several microchannel reactor units. One type, an ***integrated microchannel combustor/evaporator***, is used to generate heat by using the heat of reaction obtained through the gas-phase combustion of methane or hydrogen. For experimental purposed, the fluid receiving the heat has been water. A typical methane/oxygen reactor unit is pictures in Figure 6. Heat flux through the integrated heat exchanger has reached as high as 60 watts/cm^2. Overall system efficiencies have ranged between 85% and 93% (based on the higher heating value of the reactant gas) at operations producing 25 watts/cm^2. Methane/air units have additionally been fabricated, with slightly lower heat fluxes and system efficiencies.

PNNL researchers have additionally investigated ***catalytic microchannel reactors***, depicted in Figure 7, both to serve as heat sources and for general chemical processing. In the former case, heat fluxes exceeding 20 watts/cm^2 were achieved for integrated microchannel reactor/evaporator units with conversions in excess of 90%.

PNNL investigations of catalytic reactors have included ***partial oxidation microchannel reactors***, for methanol, butane and methane. The test unit, show in Figure 8, consists of individually fabricated microchannel heat exchangers and reactors. For the butane partial oxidation tests, higher temperatures and excess oxygen were needed to reduce coking. At 700 C, essentially 100% conversion and 30-35% selectivity to hydrogen were obtained with no observable coking. With no excess oxygen added, greater than 90% selectivity to hydrogen was achieved initially, but coking resulted in a gradual rise in the pressure drop and decrease in the selectivity.

PNNL researchers have also fabricated low-temperature, micromachined plasma reactors, incorporating channels having cross-sections of about one millimeter by one millimeter (not quite microchannels), and an overall length of about four centimeters. The units were formed in a machinable ceramic (MACOR), with defect-free dielectric materials being added using photolithographic/sputtering techniques. In proof-of-principle experiments, the micromachined units were observed to produce methane activation products approximately four times higher than in a companion non-micromachined plasma reactor (that occupied 1000 times the volume) while being operated at the same flow rates. In addition, the micromachined unit required only ¼ of the power that was consumed by the larger system.

System Architectures and Applications

As previously stated, microsystem architectures are needed that will enable large numbers of microchemical components to collectively sum their products. An example of such a system is shown in Figure 10. In this depiction, layers are assembled containing arrays of microcomponents, with each layer performing one or more unit operations. Collectively, the units would provide a complete chemical process. Potential near-term versions of these systems include hybrids of conventional technology and microtechnology, combining “macro” components such as conventional pumps and valves with micro chemical reactors and separators.

The investigations t PNNL include the development of several systems, for both energy and chemical applications. For example, all of the microcomponents of an absorption cycle heat pump, wich incorporates a heat-actuated, thermochemical compressor, have been demonstrated. The thermochemical compressor, as depicted in Figure 11, consists of a microchannel gas absorber, a microchannel heat exchanger operated in a recouperative mode, a small (but not microscale) pump, and a microchannel gas desorber. Combined with microchannel heat exchangers acting as the condensing and evaporating units, the system that we expect to demonstrate this year should constitute a compact, ***microtechnology-based heat pump***. Possible applications include man-portable cooling and environmental conditioning for automobiles.

Fabrication Techniques

A number of different fabrication methods have been used to create high aspect ratio microchannel structures at PNNL. Fabrication techniques used at PNNL have included conventional machining (slitting saw), photolithography, non-reactive ion etching and chemical etching, electrodischarge machining (EDM), and photoablation by excimer laser. Materials include copper, aluminum, stainless steel, high temperature alloys, plastics and ceramics.

Conclusions

Microsystem technologies offer the opportunity for the development of a new class of chemical process systems. Although a number of significant research issues must be favorably resolved, it should soon be possible to fabricate complex chemical systems within hareware volumes that are measured in cubic centimeters. Alternately, the microchannel components may be assembled into larger system architectures, at approximately the same size as a personal computer or slightly larger, which can provide comples chemical conversions and/or separations at high production rates.

Acknowledgements

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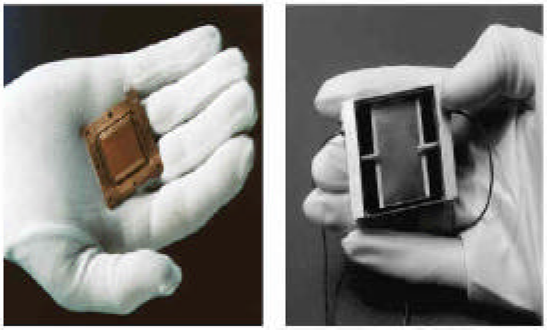
Figure 1
Simple Microchannel Array Consisting of Two Header Areas and Nineteen Flow Paths (Microchannels)



Figures 2 and 3
Microchannel Heat Exchangers

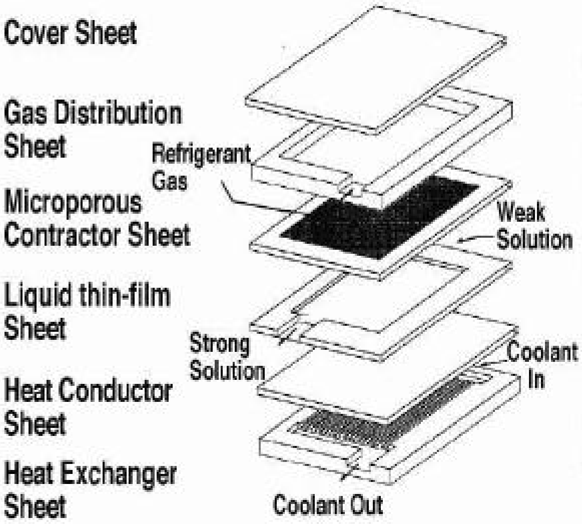


Figure 4
Microchannel Gas Absorber

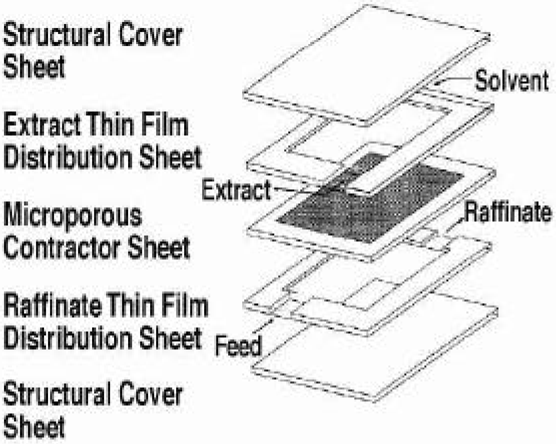


Figure 5
Microchannel Solvent Extraction Contractor

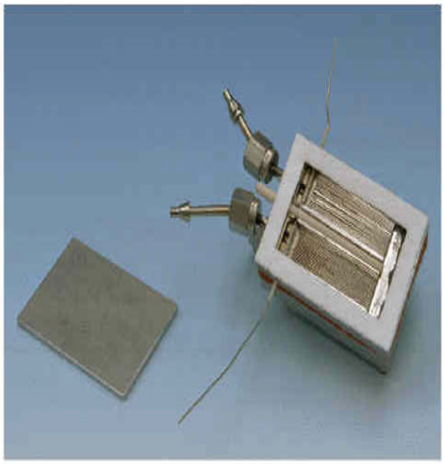


Figure 6
Gas-Phase, Integrated Microchannel Reactor/Evaporator

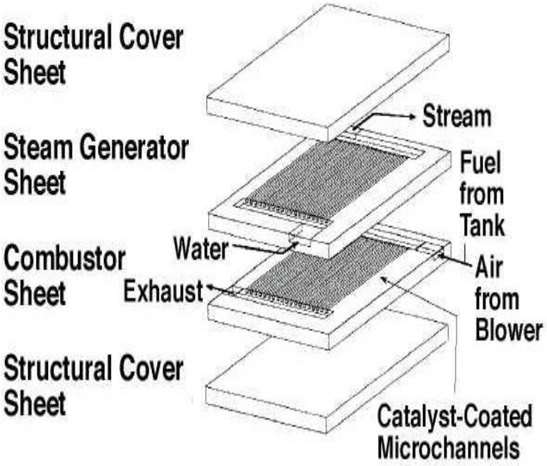


Figure 7
Catalytic Microchannel Reactor Schematic

Figure 8a
Catalytic Partial Oxidation Reactor-Components

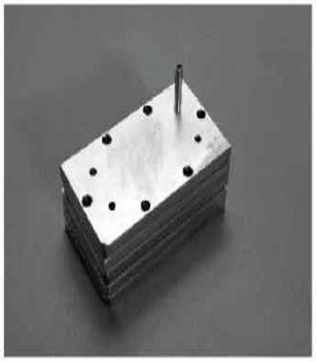


Figure 8b
Assembled Test Unit

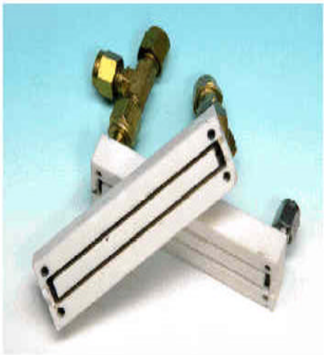
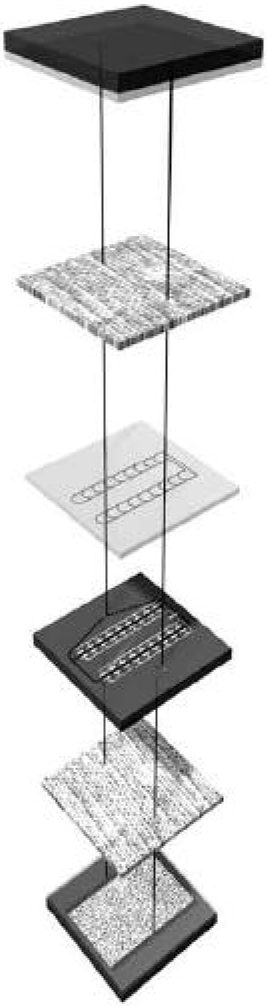


Figure 9
Low-Temperature Microchannel Plasma Reactor



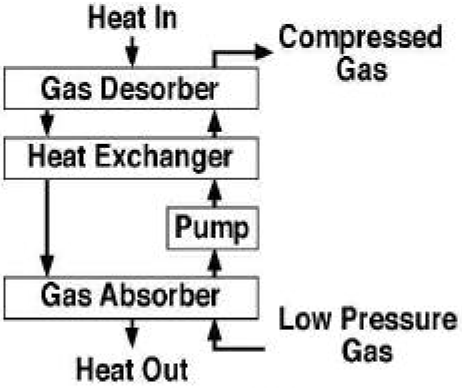


Figure 11
Diagram of Thermochemical Compressor

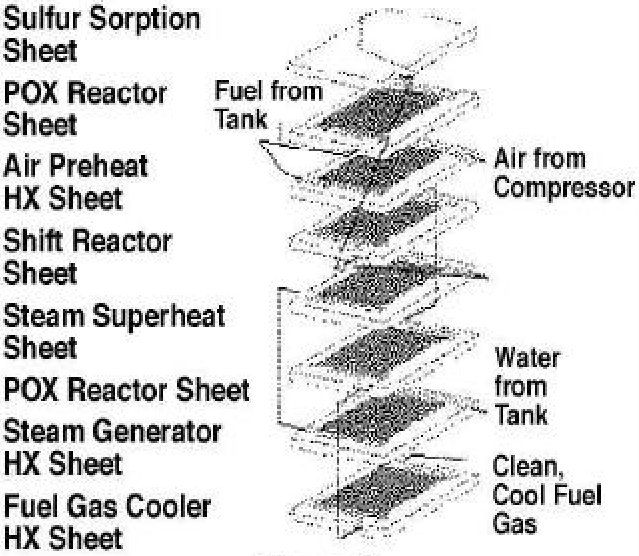


Figure 12
Microtechnology-Based Fuel Processor Schematic