Opportunities for Distributed Processing Using Micro Chemical Systems

Robert S. Wegeng and M. Kevin Drost
Pacific Northwest National Laboratory
P.O. Box 999, MS P7-41
Richland, WA 99352

As this special topical conference on process miniaturization demonstrates, work is under way at a number of research institutions to develop microfabricated components that perform the same standard unit operations that are present in large chemical processing plants, including pumps, valves, compressors, heat exchangers, chemical reactors, and chemical separations units. In the foreseeable future, we can now anticipate the assembly of compact, chemical processing and energy conversion systems that range in size from being smaller than a cubic centimeter to assemblies encompassing several cubic meters [1] [2] [3] [4] [5].

When these microsystem technologies are available, they will likely represent a new class of chemical process system, with some differences that make them significantly different from classical process technology. First, they will be able to exploit microscale phenomena more directly and more effectively than conventional technologies do. In some cases, this includes the extremely rapid heat and mass transport that is available through engineered microstructures, thus yielding components with high processing rates. Secondly, they should be mass producible, therefore at least partially offsetting economies of scale that are routinely achieved only through the operation of large chemical processing plants. Finally, the combination of the above two items plus their relatively small sizes (compared to large chemical plants) will allow them to be deployed in distributed processing applications.

While it is by no means certain what their ultimate potential is, we can now speculate regarding a number of applications where compact technologies based on microsystems might be useful. It is the purpose of this paper to briefly list and describe a few of these possible applications.

DISTRIBUTED HEAT PUMPS

In previous papers, we have discussed our work to develop microtechnology-based heat pumps, including our concept for a “sheet heat pump.” [6] [7] We are also working on a manportable heat pump for which the market already exists: cooling for soldiers in biohazardous conditions.

More generally, we note that one of the most difficult threats for military organizations to deal with today is biohazardous weapons. Accordingly, there is now a need for two types of equipment that micro chemical systems may ultimately provide. One type is a biohazard detector that can provide an accurate and reliable assessment of airborne pathogens. Another type is a lightweight, manportable cooling unit that the soldier can utilize when attired in airtight, protective clothing.

In this conference, the audience will undoubtedly hear about both needs. The development of biopathogen detectors using micro chemical components is a separate research program at. In addition, a paper is being given at this conference by researchers at the Pacific Northwest National Laboratory (PNNL), describing the development status of a compact, heat-actuated heat pump [8]. Although the unit is for energy conversion, and not for chemical processing and manufacturing, it is of interest because it includes micro chemical components that perform unit operations of interest to chemical engineers. These include microchannel heat exchangers plus an assembly of components making up a thermochemical compressor. The components in this unit include a microchannel gas absorber and a microchannel gas desorber.

High performance of this compact heat pump system is made possible because of the rapid heat and mass transport available in microchannels, and the system's ratio of cooling to hardware volume is expected to be 60 times what is routinely achieved in industrial absorption cycle heat pumps. The manportable version of this system would be run through the combustion of a liquid hydrocarbon fuel; the total system weight, including the fuel, for the manportable system is estimated to be less than ten pounds.

If this unit is successfully developed, logical spin-off applications include civilian use for a number of purposes, including cooling for emergency workers (e.g., firefighters), and embedded cooling units for shipping containers. Slightly larger versions of this system may eventually find its way into automobiles and commercial or residential structures.

DISTRIBUTED, MOBILE AND MANPORTABLE POWER GENERATION SYSTEMS

At least five papers in this special topical conference cover the development of heterogeneous, catalytic microchannel chemical reactors. These units in principle take advantage of both the heat and mass transport effects in microchannels, and in some cases therefore will be expected to yield compact reaction systems with very high throughputs[9]. Exceptions will be cases where the reaction of interest has kinetics that are very slow.

One application is the automobile. As has been reported in recent news articles, a number of automotive manufacturers now plan to introduce automobiles into the market, perhaps as early as by the year 2003, that include fuel cells as replacements to the internal combustion engine. These vehicles would still carry liquid hydrocarbon fuels, plus a key additional item: An onboard fuel processing plant to produce hydrogen for the fuel cell from the fuel source.

The Proton Exchange Membrane (PEM) fuel cell is the baseline electricity generator for these plans. The baseline system being developed by the U.S. Department of Energy (in concert with the U.S. automotive manufacturers as part of their Partnership for the Next Generation of Vehicles [PNGV]) includes reactors to perform partial oxidation reactions, to produce syngas, water-gas-shift reactions to convert the energy content of the carbon monoxide in syngas to hydrogen, and preferential oxidation reactions to reduce CO levels to less than 10 ppm, so that it will not poison the fuel cell catalyst.

A key consideration is the volume/weight and the efficiency of the fuel processing/fuel cell system. Effective thermal management is needed, especially where exothermic reactions are used, to ensure that waste energy is recycled within the process. As is reported in other papers within this conference, progress in the development of catalytic microchannel reactors leads us now to believe that an automotive fuel processing system, including all reactors and heat exchangers, would be less than 8 liters in volume (e.g., less than 0.3 cubic feet [10]. As the PNGV target is more than ten times this size, it appears likely that the microchannel fuel processing system is a candidate for inclusion in fuel-cell powered automobiles.

The technology can likewise be modularized, for both very small systems and larger systems. The distributed power generation market is one logical area, with fuel cell units and fuel processing units together converting the energy content of natural gas to megawatt scale electricity.

Likewise, the successful development of the technology would enable manportable power generation. Our expectation is that a manportable power generation system for military applications would weigh less than two pounds. Of interest, a further weight savings/efficiency savings is provided if the power system is integrated with the heat-actuated, manportable cooling unit mentioned above, with waste heat from the fuel processing system driving the thermochemical compressor.

COMPACT CLEANUP UNITS FOR WASTE TREATMENT

Liquid-liquid separations, based on solvent extraction in microchannels, is the subject of at least two papers in this special topical conference [11] [12]. While there may be a number of industrial applications for microcomponents performing this unit operation, we will proceed to discuss what may be one of the most difficult possible applications for micro chemical systems. However, if we are successful, the prize will be significant.

First, however, some history is in order. Our original interest in developing compact chemical processing systems based on microfabricated components began several years ago as a possible consideration for the remediation of environmental problems at Department of Energy sites. As the members of this audience may already be aware, cold war activities during the 1950s through the 1970s resulted in a number of DOE sites having radiochemical wastes remaining in underground storage tanks, and also resulted in a number of contaminated soil sites. An addition, a recent item in the news has been the realization that some of the tank wastes have now reached groundwater at the Hanford site near Richland, Washington.

The tank wastes are particularly troublesome. At Hanford, there are 177 underground storage tanks containing several tens of millions of gallons of material to be dealt with. The wastes consist generally of sludges, salt cake, and aqueous materials containing transuranics and fission products plus other materials remaining from the radiochemical processing of spent nuclear fuel. While there is a large volume of material overall, most of the tanks contain only a small mass fraction of radioisotopes that require the type of societal isolation that could be accomplished in deep geologic repositories. Accordingly, extensive chemical separations have often been discussed for these wastes, to reduce high level radioactive waste volumes.

The current baseline for Hanford tank waste cleanup, however, includes only a modest degree of separations, resulting in high volumes of waste for the geologic repository. One reason for this is the cost of radiochemical processing facilities, which typically require seismically hardened facilities with special HVAC equipment. To accomplish economies of scale, large radiochemical processing facilities are often envisioned, with capital costs exceeding $1 million and lead times for design and construction exceeding ten years.

Another reason is technological and economic risk. There are significant differences between types of radiochemical tank wastes, and there is a great deal of uncertainty regarding the processes that may successfully process these wastes. This translates to the possibility that a large radiochemical facility might be unable to fully process some or much of the materials for which it would be designed.

An alternative approach that we are investigating is the processing of tank waste in modular units incorporating micro chemical systems. If we are successful, the prize is the ability to assemble individual, compact chemical separations systems for each type of waste, providing extensive radiochemical separations and therefore yielding a low volume material for further treatment and ultimate disposal in the geologic repository. If we are very successful, the units might be small enough to be lowered into a tank through a riser. The result would be a large economic savings to the taxpayer, likely ranging in the billions of dollars.

It should be noted that we are not very close yet in achieving this goal. However, we are encouraged by the progress in our laboratories at PNNL, and in the laboratories of our colleagues in England, at the British Nuclear Fuels Ltd. and CRL laboratories. In this case, the rapid mass transport available in microchannels could take reduce contact time requirements for solvent extraction down to considerably less than a second. However, there are a number of other issues to be addressed if this technology is to be applied to radiochemical tank wastes. In the interim, the hardware development activities may well lead to compact separations system that could provide downwell treatment of groundwater, remove metals from aqueous wastes on ships, or provide a number of industrial services.

CHEMICAL PROCESSING ON MARS

Though it is a somewhat less hazardous environment than the interior of Hanford radiochemical tanks, Mars is nevertheless one focal point for some activities developing compact chemical processing plants. In this case, the interest is reducing the costs associated with robotic and human missions to Mars. NASA has determined that this can be accomplished by substituting process technology for propellants and oxygen for the return voyage.

The question of life on Mars has recently been highlighted by the discovery of fossil-like structures on a Martian meteorite, increasing the support for robotic sample return missions and perhaps for a human mission. A significant portion of the cost of such missions is the cost of delivering and transporting materials into and through space. NASA's plan is to include lightweight, chemical systems as part of its Mission to Mars program, to process indigenous space materials to produce propellants, oxygen and other chemicals. The program uses a phased approach, with the following elements:

Robotic Missions		
Mars ISPP Precursor (MIP) mission	2001	Demonstrates small scale O2 production
Mars PUMPP	2003	ISPP Utilization Demonstration
Mars Sample Return mission	2005	Utilizes ISPP propellants for propulsion
Mars Sample Return mission	2007	Produces and utilizes ISPP propellant and O2
Manned Mission		
Human Return Predeploy	2011	Propellants and oxygen are produced before crew departs
Earth		
Crew Launches	2013	

The NASA program for the development of compact chemical processing systems is part of its Insitu Resource Utilization Program (ISRU), which is managed by the Johnson Space Center in Houston, Texas. As part of the ISRU Program, NASA is working with PNNL to development micro chemical systems for these applications.

On Mars, the ISRU Program goal is to process atmospheric gases, which are predominately CO_2, reacting them with stored hydrogen from Earth, to make propellants and oxygen. An example of a microtechnology-based system could consist of microchannel adsorption units (for the acquisition of atmospheric CO_2), an electrochemical unit (for the reduction of CO_2 and to separate O_2), a catalytic microchannel reactor (for methanol synthesis from CO and H_2), and a microchannel liquid-vapor separator (for the separation of methanol from unreacted CO and H_2). Embedded microchannel heat exchangers may be able to provide for thermal integration of the system, for example, allowing the exothermic heat from one unit to support the heat requirements of another. Several process flow sheets are being considered, with other reactions of interest including reverse-water-gas shift, the Sebatier Process, and other Fischer-Tropsch syntheses.

DISTRIBUTED SYSTEMS FOR GLOBAL CARBON MANAGEMENT

There is a growing international concern that continued increases in atmospheric greenhouse gas levels could lead to, or may already be driving, a warming of the surface temperatures of Earth. The gas of greatest concern here is CO_2, which is primarily produced through the use of fossil fuels. While there is great uncertainty regarding the effects of global warming, the international community has agreed that atmospheric CO_2 levels must be stabilized, and the President of the United States of America has called for the development of technologies that would lead to CO_2 emission reductions.

CO_2 sequestration concepts in discussion include isolation in the same (or other) geologic strata from whence the fossil fuel originated (on land or under the ocean) or in the deep ocean where pressures cause the CO_2 to assume a form where its density is greater than that of water. Most systems that are discussed for CO_2 capture involve the installation of gas absorption equipment at central, fossil fuel power plants. In these cases, their operation would reduce the efficiency of the power generation system, requiring an increase in electricity costs. In general, carbon management strategies that are in discussion result in significant economic penalties.

Distributed processing systems, based on micro chemical systems, may have significant advantages, especially if combined with another modular technology: Fuel Cells. For example, distributed fuel cells combined with microchannel fuel processing systems can be used to produce electricity from natural gas or other fuels. Because fuel cells are not carnot-cycle limited, systems based on fuel processing and fuel cells can be inherently more efficient than internal combustion or steam power plants. In addition, modified versions of the equipment for CO_2 acquisition on Mars may be amenable to integration with the fuel processing system. Because this hardware is heat-actuated, in principle it could be run off the waste heat from the fuel processing system. This is particularly true if the primary conversion reactor is a partial oxidation unit. Researchers at Minnesota State University previously have demonstrated the catalytic, partial oxidation of methane [13], and PNNL work on catalytic, microchannel reactors for this purpose is discussed in another paper at this conference [14].

Well-head processing is another location that micro chemical systems could be valuable for the reduction of CO_2 emissions. For example, it would be useful to be able to produce high hydrogen content fuels (e.g., H_2 or methanol), and capture CO_2, on an offshore platform. Currently, some natural gas is flared off at these locations due to poor economics associated with transport of gaseous fuels, and CO_2 is typically vented from such locations as well. Conventional technology that could fit on an offshore platform cannot process hydrocarbons at the required rates; however, we estimate that micro chemical systems could be developed that may be able to accomplish this. Again, much additional research and development is necessary if this is to be accomplished, but it would be very valuable if it is determined that carbon sequestration is required to reduce global warming.

CONCLUSIONS

At the beginning of this paper, the comment was made that it is now conceivable that chemical processing systems will be manufactured, based on microsystems technology, that are extremely compact, with sizes as small (or smaller than) a cubic centimeter, or assembled in structures that take up several cubic meters. Some of these systems will have extremely high processing rates (especially when viewed as processing rate per hardware volume).

Analogies to computers are extremely tempting. Systems at the smaller end of this scale can be thought of as being analogous to computer chips and handheld calculators, as they will be extremely modular and portable. The manportable heat pump that we discussed fits within this size range. Likewise, the technologies for the production of propellants and oxygen on Mars may fit within this size regime.

Systems in the middle of this scale can be thought of as being analogous to notebook and desktop computers, with a lesser degree of portability but nevertheless being readily amenable to distributed processing applications. Here we presented automotive fuel processors and environmental cleanup units as possible examples of these. And finally, systems at the larger end of this scale can be thought of as being analogous to super computers, capable of high processing rates. To this end, we presented the possibility of hydrocarbon processing units for application on offshore platforms, which could help reduce the threat of global warming.

In all cases, a significant degree of research and development is still required. Research topics include microfluidics, materials, chemistry, reliability, and manufacturing techniques. However, the current progress convinces the authors that the concepts are fundamentally feasible. If the vision is realized, a new class of process system will be enabled.

In closing, we note that our generation has seen the development of microelectronics. When information processing became extremely fast, easy to use, inexpensive and readily available for distributed application, our society found an incredible number of applications. Accordingly, the authors pose the following question to the audience: In the future, how will we use energy and chemicals when process technology is compact and lightweight, with relatively low capital costs (through mass production) and available for distributed processing applications?

LITERATURE CITED

LITERATURE CITED

[1] Wegeng, R., Call, C. and Drost, M., "Chemical System Miniaturization," PNNL SA-27317, *AICHE 1996 Spring National Meeting*, New Orleans, Louisiana, February 1996. Also available on the world wide web at: **Error! Bookmark not defined.**

[2] Ehrfeld, W., V. Hessel, H. Mobius, Th. Richter,, V. Hessel, and K. Russow, "Potentials and Realizations of *Workshop on Microsystem Technology for Chemical and Biological Microreactors*, February 20-21, 1995.

[3] Ehrfeld, W., Cl. Gartner, K. Golbig, V. Hessel, R. Konrad, H. Lowe, Th., Richter, and Ch. Shulz, "Fabrication of Components and Systems for Chemical and Biological Microreactors," *Microreaction Technology – Proceedings of the First International Conference on Microreaction Technology*, February 1997.

[4] Lerou, J., M. Harold, J. Ryley, T. Ashmead, C. O'Brien, M. Johnson, J. Perrotto, C. Blaisdell, T. Rensi, and J. Nyquist, "Microfabricated Minichemical Systems: Technical Feasibility," proceedings of the DECHEMA *Workshop on Microsystem Technology for Chemical and Biological Microreactors*, February 20-21, 1995.

[5] Wegeng, R., Drost, M., Ameel, T. and Warrington, R., "Energy Systems Miniaturization Technologies, Devices, and Systems." Proceedings of the International Symposium on Advanced Energy Conversion Systems and Related Technologies, RAN 95, December 1995.

[6] Drost, M.K., C.J. Call, J.M. Cuta and R.S. Wegeng,, "Microchannel Integrated Evaporator/Combustor Thermal Processes", Journal of Microscale Thermophysics Engineering, 1977. Vol 1, No 4, pp 321-333

[7] Drost, M.K. and M.F. Friedrich, 1997, "Miniature Heat Pumps for Portable and Distributed Space Conditioning Applications" presented at 1997 Intersociety Energy Conversion Engineering Conference.

[8] Friedrich, M. and M. Drost, "A Microtechnology-Based Chemical Heat Pump," Joint AICHE-DECHEMA Special Topical Meeting: Process Miniaturization: Second International Conference on Microreaction Technology, March 1998.

[9] Tonkovich, A., C. Call, D. Jimenez, R. Wegeng, and M. Drost, "Microchannel Heat Exchangers for Chemical Reactors *Heat Transfer – Houston 1996 (AICHE Symposium Series 310)*, pp. 119-125.

---

[10] Tonkovich, A., J. Zilka, D. Jimenez, M. Lamont, and R. Wegeng, "Microchannel Chemical Reactors for Fuel Processing," Joint AICHE/DECHEMA Special Topical Meeting*: Process Miniaturization – Second International Conference on Microreaction Technology*, March 1998.

[11] Shaw, J., C. Turner, B. Miller and I. Robins, "Characterization of Micro-Contactors for Solute Transfer between Immiscible Liquids and Development of Arrays for High Throughput," Joint AICHE/DECHEMA Special Topical Meeting*: Process Miniaturization – Second International Conference on Microreaction Technology*, March 1998.

[12] TeGrotenhuis, W., R. Cameron and R. Wegeng, "Micromachined Contactors for Liquid-Liquid Separations," Joint AICHE/DECHEMA Special Topical Meeting*: Process Miniaturization – Second International Conference on Microreaction Technology*, March 1998.

[13] Hickman, D. and L. Schmidt, "Production of Syngas by Direct Catalytic Oxidation of Methane," *Science*, Vol 259, p. 343, 1993.

[14] Tonkovich, A., Call, C., and Zilka, J., "The Catalytic Partial Oxidation of Methane in a Microchannel Chemical Reactor," Joint AICHE/DECHEMA Special Topical Meeting*: Process Miniaturization – Second International Conference on Microreaction Technology*, March 1998.