Process Intensification Through Miniaturization of Chemical and Thermal Systems in the 21st Century

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Abstract

The 21st century holds great promise for development of Micro Chemical and Thermal Systems (MICRO-CATS™). The quest for miniaturization will lead to greater process intensification. Miniaturization greatly reduces the resistances to heat and mass transfer. When heat and mass transfer rates are increased by orders of magnitude, revolutionary changes occur in technology.

Miniaturization of electronics over the past few decades has transformed the way we live. The invention of the transistor meant that small, lightweight portable radios could be carried anywhere. Not long ago the idea of chips was revolutionary; today chips make hand-held computers, cell phones, and many other hand-held electrical devices possible. These are examples of distributed technology made possible by miniaturization.

Miniaturization of chemical and thermal systems will take us on a similar technological journey. Already demands are driving the development of MICRO-CATS. The quest for miniature systems is leading to man-portable cooling, automotive fuel processing, and in situ resource utilization for space exploration. The 21st century will see great progress in the development and use of engineered nanosystems. In electronics, the concept of "molecular electronics" is seen as a way to further increase the power. The same will occur in MICRO-CATS. We envision using engineered nanosystems to create more powerful microscale devices with highly functional surfaces that incorporate enzymes and chemical catalysts. These surfaces will make the device or system more efficient and reliable; they will be able to repair, renew, or replace themselves; eliminate or heal themselves from surface fouling; and carry out step-by-step chemical reactions. Molecular coatings will create a molecular assembly line. By incorporating enzymes, reactions that are conventionally carried out at high temperatures and pressures can be achieved at ambient conditions. Finally, in the 21st century, the missions for outer space will create opportunities for MICRO-CATS to be major parts of the successful exploration of space. The future for MICRO-CATS is as wide open as outer space itself!

Keywords: miniaturization, microscale systems, reactors, heat pumps

1 Process Intensification

Micro Chemical and Thermal Systems (MICRO-CATS) use miniaturization to achieve process intensification. Process intensification is a measure of the amount of productivity that can be accomplished per unit volume. This increase in process intensification occurs because miniaturization significantly reduces the resistances to heat and mass transfer. Imagine heat and mass transfer rates increased by factors of 10 to 100 or more. Furthermore, when many components and functions are combined and integrated into one miniature system, good things happen. In fact, things happen that are almost too good to be true! That is where the age of miniaturization in MICRO-CATS is heading in the 21st century. Process intensification opens the way for revolutionary changes. Many things we can think of today and some things we haven't yet imagined will happen in the 21st century. For instance,

- Today, we adjust the room thermostat. Tomorrow, our clothing will be manufactured with microsystems that have the capability to automatically heat and cool us.
- Today, internal combustion engines move us around. Tomorrow, microsystems will allow different fuels and power systems to be used in our vehicles.
- Today, we launch almost everything that is needed on a space mission. Tomorrow, we “will live off the land” as we explore the universe. Microsystems will harvest raw materials and process them into what we need.
- Today, we are able to transplant some organs. Tomorrow, we will implant artificial organs composed of microsystems.

As this conference on process miniaturization demonstrates, work is under way at many research institutions and industries to fabricate microcomponents and systems that perform the same standard unit operations that are present in large chemical processing plants. Although miniaturization is in its early stages, it is driven by considerable incentive and the opportunity to revolutionize heat exchangers, heat pumps, reactors, gas absorbers, solvent extractors, fuel processors, pumps, valves, compressors, and other similar components and systems. In the foreseeable future, we anticipate the assembly of compact, chemical processing and energy conversion systems that range from smaller than a cubic centimeter to several cubic meters in size [1, 2, 3, 4, 5].

The quest for miniaturization is marching forward. The changes that have occurred in electronics in the 20th century enable us to envision what to expect from micro chemical and thermal systems in the 21st century. The advances in microelectronics have transformed computers, communication systems, and appliances, to name a few. The transistor is now about 50 years old. This invention, along with other innovations, provided for process intensification. The transistor radio changed things because it was small and light enough to carry anywhere. This created possibilities we didn’t have before.

Miniaturization of components on computer chips has made it possible to have powerful hand-held computers, cellular telephones, global positioning systems, and many other electrical devices. In fact, the telephone booth may in one more generation be found only in museums. Everyone will have compact, low cost personal cell phones or something even better. A few years ago the idea of putting so many components and functions on one microchip was revolutionary. Today, chips are an integral part of almost every electrical device.

The advances in microelectronics spurred the development of microelectro mechanical systems (MEMS). Scientists working in this field have developed silicon actuators/levers, gears and gear reduction units, diaphragm pressure sensors, motors, transmissions, locks, mirrors, hinges, and more. Some of these devices have been combined to yield intricate mechanical systems-on-a-chip. Efforts are moving toward integrated microelectronic/micromechanical systems-on-a-chip; the acceleration sensor for deploying air bags is an example of the commercialization of micromachined systems-on-a-chip.

The strategy of miniaturized distributed systems is powerful; the potential impact worldwide is enormous. For example, this is probably one of the most significant strategies for the eradication of poverty. Until now, the countries with the resources, large-scale technologies and infrastructure, and skilled labor enjoyed the highest standards of living. But distributed processing can change that equation. For instance, some third-world countries will have communications without telephone poles and underground cables. Microtechnology presents new possibilities for poor countries to increase their standard of living using distributed micro chemical and thermal systems. With these technologies, all countries can have higher productivity, power, and communications anywhere, without needing to build expensive infrastructures. So miniaturization brings with it new hope for the 21st century.

2 Evidence for Success

The evidence for success of MICRO-CATS can be found in their demonstrated performance. They have higher heat transfer rates and faster reaction rates and can be operated with lower pressure drops than conventional systems. Some of the successful results for microreactors and solvent extraction are presented below.

Researchers at Pacific Northwest National Laboratory (PNNL) have investigated several microchannel reactor units. One, an integrated microchannel stainless steel combustor/evaporator, generates heat using the heat of reaction obtained from the gas-phase combustion of methane or hydrogen [6]. The technical objective is to demonstrate that a microchannel combustor can produce at least 25 watts of thermal energy per square centimeter of heat transfer profile area and transfer that energy to a cooling unit. This heat transfer rate is approximately 20 times higher than that of a conventional water heater. The most important problem for the microcombustors is maintaining the flame in a small volume. There are also problems maintaining the desired temperature in the combustion chamber and efficiently removing the heat and the combustion products.

The overall size of the system is 41 x 60 x 20 mm. Tests were conducted on micromachined combustor channels 300 microns wide, 500 microns deep, and 35 mm long; results showed that the system achieved heat fluxes greater than 30 W/cm^2 and combustion efficiencies greater than 85%. (Combustion efficiency is the ratio of the heat transferred to the water relative to the heating value of the fuel.) In this system, oxygen (or air) and fuel were introduced into the combustion chamber where an ignition wire initiated combustion. Microchannels were fabricated on the under side of the burner to provide cooling during the combustion process. The combustion products then entered a microchannel heat exchanger that enhances heat transfer between the combustion products and the combustor solid surfaces. Flame quenching problems increased as the combustor size decreased, indicating that flame quenching will set a limit on the degree of miniaturization possible in this type of combustor.

PNNL researchers have also investigated catalytic microchannel reactors made from high-temperature metal alloys for several applications. Engineered catalysts provide large surface areas, high heat-transfer rates, and low pressure drops. While slow kinetics is a current paradigm in conventional reactor systems, fast kinetics is possible in microchannel reactors. While multisecond reactions may be necessary in conventional reactors, millisecond reactions occur in microchannel reactors. This improvement is attributed to the diffusion lengths, which are reduced by at least two orders of magnitude, and to heat transfer, which prevents “hot spots” and “cold spots” and enables high throughputs.

A methane partial oxidation reactor was operated with fast kinetics at 900°C to produce carbon monoxide and hydrogen. Methane conversion efficiencies were more than 85% with 11 millisecond residence time and 100% with 25 millisecond residence time. The use of catalysts in microchannel reactors is so promising that PNNL is now developing engineered microstructures to find those best suited to microchemical reactors.

At PNNL, we are investigating microchannel reactors using the Sabatier reaction and the reverse water-gas shift reaction. The Sabatier reaction combines carbon dioxide with hydrogen to produce methane propellant and water vapor. In the reverse water-gas shift reaction, carbon dioxide combines with hydrogen to produce more fuel and water. An electrolysis unit dissociates the water into oxygen and hydrogen. Tests showed that the Sabatier reaction catalyst provides 85% conversion at 250°C with a reactor residence time of 0.1 second. The residence time for the reverse water gas shift catalyst was even shorter. These successful results are very promising for the development of MICRO-CATS for NASA’s Mars missions.

PNNL researchers have designed and tested microchannel devices for efficient contacting of two liquids in solvent extraction [7]. Recent results are being presented by PNNL researchers at this conference [8]. Previous efforts have tested such devices as gas absorbers. This solvent-extraction work is a step in the development of compact efficient devices for chemical separations. The microchannel contactor results in substantially higher throughput per total system volume compared with conventional technologies.

Solvent extraction requires intimate contact of two immiscible liquids to facilitate mass transfer of one or more solutes from one fluid to another. The architecture in this work consists of two micromachined channels separated by a contact plate. The channels are separated by a micromachined contactor plate of 25-micron-thick Kapton substrate that has a matrix of uniform holes 25 microns in diameter. The solvent and feed streams can be operated co-currently or counter-currently. This arrangement allows for intimate contact of the two immiscible fluids as they flow through very thin channels smaller than the normal mass-transfer boundary layer.

The mass transfer resistance has contributions from each flow stream, from the contactor plate, and from the interface. The pressures inside the contactor must be carefully controlled to prevent breakthrough of the fluids through the holes in the contactor plate. If the interface is immobilized, the two liquid films can flow in counter-current directions, allowing for a more effective separation in the contactor. PNNL researchers are investigating reducing the overall size of the equipment and improve operating efficiency by reducing the thickness of the films and improving mass transfer efficiency in the contactor plate. Tests were conducted using microcontactor channels 1 cm wide, 10 cm long and 200 to 500 microns high. Results with cyclohexane-water feed and cyclohexane solvent indicate that the micromachined contact plates worked at least as well as commercial microporous membranes and have significant potential for improved performance. Also, with channel heights of less than 300 microns, diffusion through the contactor plate is the limiting factor in mass transfer.

Further evidence for success is our ability to model chemical and thermal processes at the microscale. PNNL researchers have developed a lattice-Boltzmann simulation capability for modeling the behavior of microscale fluidic systems. In fact, this work is being presented in a paper at this conference [9]. Complex fluid dynamics problems in this size range are not now amenable to being modeled by conventional simulation methods, which do not properly account for the importance of surface forces at fluid-gas, fluid-fluid, and fluid-solid interfaces. However, our lattice-Boltzmann modeling capability overcomes these limitations. This is critically important for supporting the design and testing of MICRO-CATS.

3 Progress Toward the Future

The quest for micro chemical and thermal systems is being driven by some important needs. One is the need for miniature analytical systems for DNA, toxic chemicals, and pathogens such as HIV. Such instruments are needed in the fields of medicine, forensics, agriculture, and environmental control. Other prominent needs driving development include 1) military requirements for human-portable heating, cooling, and power generating units; 2) transportation demands for hydrogen rich streams for fuel cells; and 3) chemical processing systems capable of processing the rocks, soil, and atmosphere on planets such as Mars. These are just examples. While we cannot be absolutely certain what the potential for microtechnology might be, we can speculate about areas where the demand caused by technical problems or social needs will draw us forward. At the present at PNNL, we are pursuing major opportunities for MICRO-CATS in buildings, carbon management, environmental restoration, industrial chemical processing, military, space exploration and transportation. More details on these applications are found on our web site: http://www.pnl.gov/microcats

At PNNL, we have several systems under development but just three are described in this paper: man-portable cooling, automotive fuel processing, and in situ resource utilization for space exploration. These three technologies demonstrate the diversity of potential applications.

3.1 Man-Portable Cooling

PNNL researchers are working on microsystems to provide man-portable cooling. The market is driving this work because there is a need to cool people who work in hazardous environments such as 1) soldiers in certain field operations, 2) emergency response workers for environmental cleanup, firefighters and other emergency workers, and 3) industrial workers in hot, unforgiving environments. A cooling device is needed for use with protective clothing, and it must be compact, lightweight, and able to operate for extended periods of time. We are developing absorption heat pumps using a lithium bromide and water solution as the working fluid. As this technology is successfully developed, we envision spin-off applications with slightly larger versions for cooling shipping containers and air conditioning vehicles and even commercial or residential structures.

In this conference, a poster session is being given by PNNL researchers describing the status of compact, heat-actuated heat pump development [10] (see Figure 1). 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. It is a chemical heat pump that includes 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.

The high performance of this compact heat pump system is possible because of the rapid heat and mass transport available in microchannels. Current estimates suggest a size reduction of a factor of 60 compared with conventional heat pumps is possible. The weight of the system will be reduced by at least a factor of 2. The man-portable version of this system would operate by combustion of a liquid hydrocarbon fuel; the total system weight, including the fuel, for the man-portable system is estimated to be less than ten pounds.

3.2 Automotive Fuel Processing

Automotive fuel processing is another application where the potential exists for significant market demand for microtechnology. Some automotive manufacturers plan to introduce fuel-cell-powered automobiles into the market in the next few

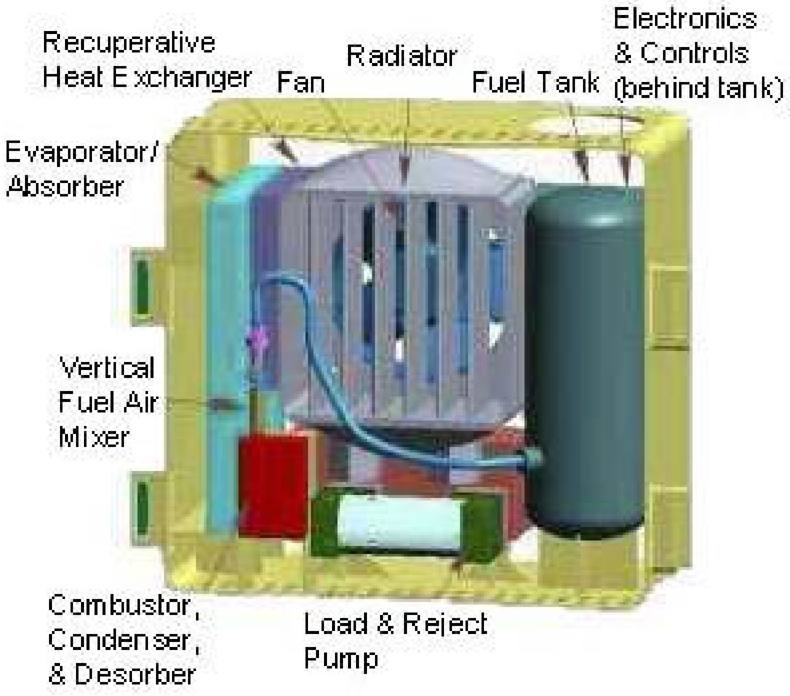

Figure 1. Mesoscopic Heat-Actuated Heat Pump

years. The fuel cells will generate electricity to drive the electric motors that move the vehicles. These vehicles will still carry liquid hydrocarbon fuels, plus a key additional item: an onboard fuel processing plant to produce hydrogen for the fuel cells.

The proton exchange membrane (PEM) fuel cell is the electricity generator for the baseline system being developed by the U.S. Department of Energy (with U.S. automotive manufacturers) in their Partnership for the Next Generation of Vehicles. The fuel processor is a critical reactor technology for the deployment of the PEM fuel cells, which operate using hydrogen. Figure 2 shows the fuel vaporizer portion of the automotive fuel processing system.

The fuel processor produces hydrogen rich streams from gasoline or methanol fuel using a heterogeneous, catalytic microchannel chemical reactor. This microreactor unit takes advantage of both the heat and mass transport effects in microchannels and is thus able to provide a compact system with high throughput. It is a multi-step process involving fuel vaporizer, primary conversion reactor to produce synthesis gas, water gas shift reactor, and CO cleanup reactor. The fuel processor has an occupied volume of less than 0.3 liters to support a 50 kWe output from a fuel cell. At this conference, PNNL researchers are presenting a paper that describes the development of this fuel processor (11).

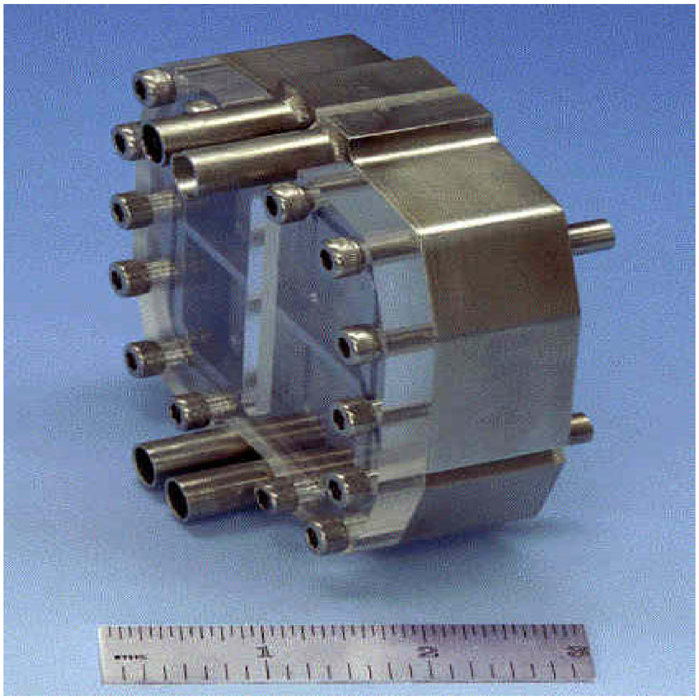

Figure 2. Microchannel Gasoline Vaporizer for 50-Kwe Fuel Processing System

3.3 In Situ Resource Utilization

Microtechnology has attractive applications for supporting space exploration and orbit-based research. For such missions, payload size and weight is of paramount importance. Compact, lightweight microsystems cost less to launch and leave room on the spacecraft for other important equipment and supplies. Before the advent of microsystems, it was not possible to carry all fuels and other chemicals from Earth and it is not cost-effective to do so now. So the strategy is to "live off the land" in space exploration.

NASA wants to use compact chemical processing plants in its Mars missions. They are interested in reducing the costs associated with robotic and human missions to Mars and have determined that this can be accomplished by substituting process technology for propellants and oxygen for the return voyage. MICRO-CATS can produce fuel and oxygen from Martian atmospheric gases for NASA astronauts during their stay on Mars and for the return trip home.

So 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 oxygen production

Mars PUMPP	2003	ISPP Utilization Demonstration
Mars Sample Return mission	2005	Utilizes ISPP propellants for propulsion
Mars Sample Return mission	2007	Produces and uses ISPP propellant and oxygen
Manned Mission		
Human Return Predeploy	2011	Propellants and O2 produced before crew departs Earth
Crew Launches	2013	

Extraterrestrial chemical processing plants will need to be compact, lightweight, efficient, and able to operate reliably for prolonged periods in reduced gravity environments. This presents unique challenges because conventional chemical process equipment relies heavily on gravity to operate effectively. If such systems are to be efficient and reliable, a fundamental understanding of operation in non-Earth environments is required.

We are working with the In Situ Resource Utilization (ISRU) program at NASA's Johnson Space Center to develop compact systems to produce fuel and oxygen from the Martian atmosphere. So far, the microchannel technologies have been conceived to convert carbon dioxide to methane and water by reacting it with hydrogen using the Sabatier process and reverse water gas shift reactions. These processes use carbon dioxide from the atmosphere to produce methane, water, and carbon monoxide. Hydrogen must be carried from earth for use in these reactions. Oxygen can be produced from the water in an electrolysis unit. The carbon monoxide can be further reacted with hydrogen to produce fuel.

4 MICRO-CATS in the 21st Century

The future is here for MICRO-CATS because many opportunities are now within our reach. We see three important fronts on the horizon of the 21st Century:

- The use of engineered nanosystems to produce "highly functional" surfaces
- The marriage of enzymes with highly functional surfaces
- Greater demands for MICRO-CATS to perform in the extreme and harsh conditions found on space missions.

4.1 Engineered Nanosystems

In the 21st century, engineered nanosystems will be used to develop MICRO-CATS. Why? Because engineered systems are often more powerful than natural ones. They present the possibility of further process intensification. Engineered nanosystems will be a platform for applying molecular sciences in the 21st century.

The ability to fabricate components is always one of the main limitations in the development of micro chemical and thermal systems. But overcoming these limitations presents opportunities for significant improvements. PNNL places major importance on developing the capability of microfabrication. Some of our progress is being presented in a paper at this conference [12]. Looking ahead, we believe that when engineered nanosystems (e.g., molecular systems) are included with microsystems, significant additional improvements in process technology may result.

Highly functional surfaces result from changes in surface properties that make the device or system more efficient, more effective, or more reliable. These surfaces may have the ability to repair, renew, or replace themselves. They could be surfaces that control or eliminate fouling—imagine a surface that can protect and heal itself from surface fouling. They could carry out a series of step-by-step chemical reactions. We envision using molecular coatings to create a molecular assembly line. In the 21st century, the use of highly functional surfaces will become routine.

Custom-designed catalysts will be developed. They will be designed and constructed to provide higher catalyst activity and greater specificity. When engineered and fabricated at the nanoscale, microsystems are more effective. This is what we can expect in the 21st century.

4.2 Enzymes and Functional Surfaces

All of life is full of microchemical systems. These have been working and developing for millions of years. Things like membranes, cells, lungs, kidneys, and hearts operate because they contain viable, effective, and reliable microtechnology. They involve chemical reactions, and virtually all of these chemical reactions require catalysts to arrange the reactants in energetically favorable orientations so that chemical products will be formed. Man-made chemical catalysts, such as platinum compounds, only crudely arrange reactants to form products, and, consequently, our chemical plants require high temperatures, pressures, and pH extremes to complete reactions. In contrast, cells require their catalysts to function under the mild conditions required for sustaining life. Nearly all reactions in cells are catalyzed by enzymes. Unlike chemical catalysts, enzymes are frequently catalytically perfect; i.e., once the enzyme binds its substrate, the reaction product is formed, sometimes at thousands of reactions per second. With the development of functional surfaces and the use of enzymes, many chemical processes that are not effected under high temperature and pressure conditions may be accomplished with enzymes and normal temperature and pressure conditions. Furthermore, the microdevices will then provide a molecular assembly line capability such that molecular changes occur in a sequenced step-by-step manner. These reactions will be dramatically different and probably dramatically better.

More than 5000 useful enzymatic reactions have been characterized to date, and many of these reactions are understood in molecular detail because the three-dimensional structures of the enzymes and reactants have been determined. DOE's genome sequencing programs and x-ray diffraction beam lines have been essential in identifying enzymes and determining their three-dimensional structures. Recombinant DNA technology makes it possible to produce virtually any enzyme in large quantities by isolating the gene, modifying the gene to enhance enzymatic properties, inserting the gene in an organism designed to express large quantities of protein, and then purifying the resulting protein. Recombinant enzymes provide attractive alternatives to constructing energy-intensive chemical plants that use crude catalysts.

A long-term goal is to someday manufacture artificial enzymes with highly functional surfaces. The structural and mechanistic insights elucidated from biological enzymes would provide the template for an advanced version of existing highly functional surfaces designed to mimic enzymes. Even if the artificial enzyme is orders of magnitude less efficient than the biological enzyme, the artificial enzyme might still be sufficient in most chemical applications and significantly less expensive than current crude catalysts.

4.3 Future in Space

The space age arrived more than 40 years ago, but it slowed when the costs became huge to carry out increasingly complex and ambitious space missions. Other priorities prevailed, and budgets for space exploration programs were not increased. But now, MICRO-CATS may help provide the way to accomplish missions at lower costs. In addition, and equally important is the fact that space exploration is one of the applications where microtechnology is better than conventional technology.

The extreme harsh environments coupled with the space mission requirements of compactness, light weight and high reliability present a quantum level change in requirements for MICRO-CATS. Can the system endure the rigors of temperature extremes? Does it operate under low-gravity or zero-gravity conditions? Can it be on stand-by for long periods and then operate reliably when needed? The rigors of developing technology for NASA and others may also spur the development for many other applications in the future.

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