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NEWS

Advanced Energy Technologies | Atmospheric Sciences | Building Technologies | Energy Analysis | Indoor Environment

An Update on the *EETD* Research Program on Batteries for Advanced Transportation Technologies

The demand for so-called hybrid-electric vehicles (HEVs), which are powered by a battery and combustion engine (or fuel cell) working in tandem, has surged during the past few years. Automakers are responding to this demand by offering more and more models with hybrid power sources. This is good news from both the environmental and energy points of view because HEVs have fewer exhaust emissions and better fuel economy than equivalent vehicles powered only by combustion engines.

The HEV's current market success is a result of its combined battery and combustion-engine configuration, which allows it to avoid many of the problems of the once-promising pure electric vehicle (EV), powered only by a battery. Environmental Energy Technologies Division (EETD) electrochemists are intimately involved in the future of both types of vehicles through the Batteries for Advanced Transportation Technologies (BATT) Program, which focuses on developing advanced high-energy rechargeable batteries for EVs as well as high-power rechargeable batteries for HEVs.

EVs *vs.* HEVS

The development and commercialization of battery-powered EVs has proven extremely challenging because EV batteries must simultaneously meet multiple, demanding requirements: high energy per unit mass (to provide acceptable driving range between battery recharges), high power (to provide acceptable vehicle acceleration and to receive energy generated during vehicle deceleration, a.k.a. regenerative braking), long lifetime, low cost, resistance to abuse and operating temperature extremes, absolute safety, and minimal environmental impact. Despite years of intensive worldwide research, no EV battery has been developed that can meet all of these goals. Perhaps the most vexing barrier is cost. There are three components to this problem: 1) advanced batteries cost between $150 and $300 or more per kilowatt hour (kWh) of stored energy, 2) a battery-powered EV must store at least 30 kWh of energy, and 3) battery lifetimes are typically shorter than five years. This combination of limitations means that there is little demand for EVs and therefore little incentive for automakers to produce and market them.

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The HEV has succeeded because it circumvents many of the EV's problems. A key advantage of the HEV is that it uses a small, e.g., one-kWh-capacity battery compared the 30 kWh or more required for an EV. This small battery significantly reduces overall vehicle cost (as detailed in "Lithium Batteries for Hybrid-Electric Vehicles," *EETD Newsletter*, Winter 2000). HEVs also avoid the recharging challenge faced by EVs because the combustion engine charges the battery while the vehicle is driven. So, although long-term research on EV batteries continues in part because of the attractiveness of EVs' zero emissions, the HEV design is currently the most viable for the mass market.

Battery Designs and Chemistries

Because HEV batteries do not need to be as large as EV batteries, HEVs can use versions of the same products that are found in today's consumer electronic devices (e.g., laptop computers, cellular telephones, power tools). The key difference between batteries used in HEVs and those used in other consumer electronics is that HEV batteries are designed to maximize *power output* per unit mass/volume, for example by using thin electrodes and heavy metallic electronic conductors. In contrast, consumer (and EV) batteries are generally designed to maximize *energy content* per unit mass/volume, for example by using thick electrodes and lighter electronic conductors.

Figure 1. Battery cell test apparatus

Today's most common HEV battery is nickel-metal hydride and uses a nickel oxide positive electrode, aqueous alkaline electrolyte, and a hydride negative electrode. The nickel-metal hydride battery meets or approaches many of requirements listed above for the EV and is sufficiently robust for HEV applications. However, the nickel-metal hydride battery has limitations such as marginal energy and power characteristics and poor ability to accept charging currents at elevated temperatures, so automakers are searching intensively for a next-generation battery system with improved performance.

Because the lithium-ion battery has emerged as the system of choice for many consumer electronics applications, it has also attracted major interest as the likely battery of the future for HEVs. Present-day lithium-ion batteries designed for consumer electronics use a carbon-based negative electrode, an organic liquid electrolyte with dissolved lithium salt, and a cobalt oxide positive electrode. However, although the cost of lithium-ion batteries has dropped significantly during the past few years, it remains expensive for HEV applications, so there is strong incentive to develop less-expensive cell components (e.g., to replace at least some of the cobalt with another metal). Lithium-ion batteries also pose a potential safety problem because of the instability and flammability of the organic electrolyte. This problem can be overcome for small consumer batteries that contain only a few Wh of stored energy, but it is a major concern for HEV batteries that contain about one kWh of stored energy.

The BATT Program

Lawrence Berkeley National Laboratory (Berkeley Lab) has had a long-standing "Lead Laboratory" role in the battery research area of the U.S. Department of Energy FreedomCAR and Vehicle Technology Program (http://berc.lbl.gov/BATT/BATT.html). BATT Program research addresses the fundamental chemical and mechanical instabilities that have impeded the development of EV batteries that have acceptable cost, lifetime, ruggedness, and safety. This work supports the development of both advanced high-energy rechargeable batteries for EVs and high-power rechargeable batteries for HEVs. BATT Program researchers assemble advanced components into battery cells, determine failure modes, synthesize and evaluate new materials, carry out advanced diagnostics, and develop sophisticated electrochemical models to improve our understanding of the complex processes that affect battery performance and life.

Baseline Battery Chemistries

An essential feature of BATT research is the selection of battery chemistries to serve as "baseline" systems against which new materials and designs can be compared. There are currently three BATT Program baseline cell chemistries; all are variants of the lithium-ion battery and include low-cost natural graphite as the negative electrode and an organic electrolyte with dissolved lithium salt. The major difference among the three is the material used for the low-cost positive electrode, usually called the cathode. The three cathode materials in the baseline systems are: mixed transition metal oxide, $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$, which provides high voltage and high energy; lithium iron phosphate, $LiFePO_4$, which provides lower voltage but excellent stability; and manganese oxide spinel, $LiMn_2O_4$, which provides high voltage and high power.

In a project led by Kathryn Striebel, baseline materials are incorporated into standardized cells and tested using a consistent protocol to determine capacity, energy, power, and lifetime characteristics (see "Testing New

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Battery Materials in Standard Cells," *EETD Newsletter*, Fall 2003). Striebel and her colleagues benchmark BATT Program baseline cells against established battery performance goals, evaluate new materials, and then disassemble the cells and components for detailed characterization to determine failure modes (see Figure 1). They also test advanced cell designs suggested by BATT modeling groups, led by John Newman and Venkat Srinivasan.

Overcharge Protection

One critical concern is developing approaches to protect batteries from overcharging. Because HEV and EV electric motors operate at 300 volts or higher and individual lithium-ion cells deliver only three or four volts each, many cells must be arranged in series to provide the needed voltage. In contrast to batteries with an aqueous electrolyte (e.g., the nickel-metal hydride batteries currently used in HEVs), which can be designed to accept limited overcharging, batteries with a non-aqueous electrolyte cannot easily be made to accept overcharging. This is a particularly serious problem for HEVs and EVs because if even one cell in the series is slightly weaker than the others, it will become overcharged unless corrective measures are taken. Overcharging of a lithium-ion battery cell is extremely dangerous because it not only heats the cell but also destabilizes cell components, and both processes can result in cell leakage, fire, and explosions. Unfortunately, established corrective measures for consumer lithium-ion batteries (e.g., individual electronic control of each cell) are expensive and cumbersome. To address this problem in a practical manner, Tom Richardson and his BATT colleagues are developing novel electro-active conducting polymers that can protect against overcharge in lithium-ion cells.

Alternative Anodes

Present-day lithium-ion batteries, which employ various forms of carbon as anode materials, suffer from safety, cycle-life, and storage-life problems. BATT Program researchers are studying non-carbonaceous anodes as possible alternatives to address these problems. Low-cost metal alloys and carbons with acceptable capacity, rate, cyclability, and calendar life are under investigation (see Figure 2). The interaction of these alloys with the organic electrolyte is complex and poorly understood but is believed to be a critical process that destabilizes and thereby limits the lifetime of alloy anodes. Philip Ross and Vera Zhuang [of the Berkeley Lab Materials Sciences Division (MSD)] and colleagues are using advanced characterization techniques to improve our understanding of how these alloy components interact with the electrolyte. This research has increased our understanding of model electrodes (i.e., very smooth thin films of a single anode component), and researchers are close to applying this knowledge to explain alloy behavior.

Safer Electrolytes

BATT Program electrolyte research, led by Nitash Balsara (MSD) and John Kerr, focuses primarily on

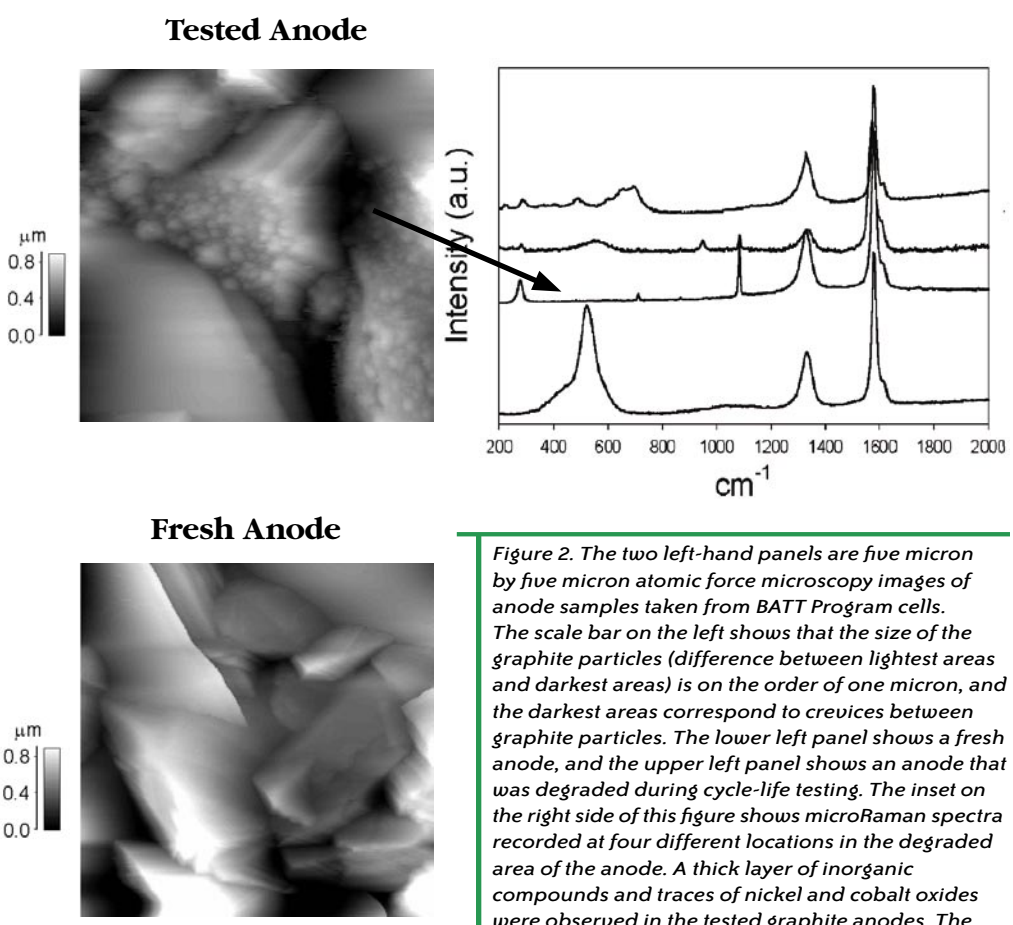

Figure 2. The two left-hand panels are five micron by five micron atomic force microscopy images of anode samples taken from BATT Program cells. The scale bar on the left shows that the size of the graphite particles (difference between lightest areas and darkest areas) is on the order of one micron, and the darkest areas correspond to crevices between graphite particles. The lower left panel shows a fresh anode, and the upper left panel shows an anode that was degraded during cycle-life testing. The inset on the right side of this figure shows microRaman spectra recorded at four different locations in the degraded area of the anode. A thick layer of inorganic compounds and traces of nickel and cobalt oxides were observed in the tested graphite anodes. The nickel and cobalt oxides probably migrated from the cathode to the anode during the cell test.

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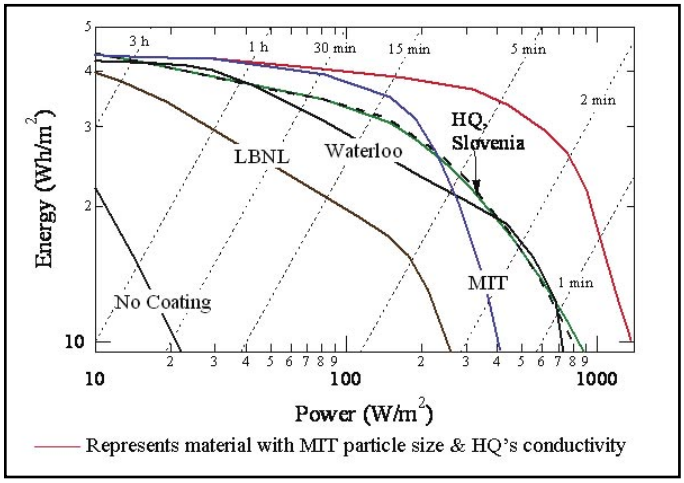

Figure 3. Results of a detailed mathematical analysis of a single, non-optimized, lithium-ion battery cathode design. Important design variables are specific loading of lithium iron phosphate, which is the electrochemically active component of this particular cathode, and cathode thickness. This modeling approach not only allows for differences in cell design to be normalized, but also captures the importance of intrinsic cathode properties such as particle size, conductivity, and carbon content, all of which vary dramatically according to the exact method of cathode preparation. Note that the solid red curve represents what could be achieved by using the particle size of one material and the conductivity of another material. The ordinate is proportional to battery power (corresponds to vehicle acceleration), and the abscissa is proportional to battery energy (corresponds to vehicle distance traveled during battery discharge).

polymer electrolytes, which are safer than liquid organic electrolytes and will not flow or leak from cells. Prior efforts (including major, multi-year, multi-million-dollar projects by industrial consortia) to develop practical lithium batteries based on polymer electrolytes have been hampered by the polymer electrolyte's low ionic conductivity and the critical problem of lithium metal dendrites, which are tree-like branched structures that grow across the electrolyte and short the electrodes. Resolution of these two problems is recognized as an extremely difficult task. BATT Program scientists searching for polymer electrolytes with improved properties are studying composite polymer-particle mixtures, diblock polymers with aligned cylindrical structures, oligomeric anions, and other unusual assemblies.

New Cathode Materials

The identification of novel cathodes is critical because of the high cost and environmental problems of the cobalt oxide materials used in today's lithium-ion batteries. An important BATT goal is to develop a high-rate stable manganese-based cathode. Although manganese is a low-cost constituent, manganese-based cathodes tend to lose capacity at unacceptable rates. BATT research is directed at understanding the reasons for this deterioration in capacity as well as evaluating novel forms of manganese-based cathodes that exhibit high capacity (greater than 150 ampere-hour per kilogram of oxide) and acceptable stability.

Two of the baseline cathode materials mentioned above are examples. The mixed transition metal oxide $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ has a layered structure into which a large amount lithium can be inserted, and it has the potential for higher coulombic capacity and higher energy than cobalt oxide, at a lower cost. The manganese oxide spinel $LiMn_2O_4$ has a three-dimensional structure that offers an easy path for inserting lithium ions and thereby supports high ion-transport rates and high battery power. Marca Doeff (MSD) and Tom Richardson are leading the Berkeley Lab effort to develop improved cathode materials for the BATT Program.

Diagnostics and Modeling

Advanced diagnostics are essential to investigating the processes that limit battery life and performance. Unfortunately, it is either impossible or very difficult to use most of the common analytical tools to characterize the specific processes that affect battery life and performance. The reasons for this are varied and complex, e.g.:

- battery charge and discharge involve numerous simultaneous and inter-related phenomena, including ion transport, electron transport, molecular transport, chemical reactions, electrochemical reactions;
- degradation processes may constitute only a small fraction of the electrochemical process and may require many months or years to become apparent;
- there is no accepted "accelerated life test" for batteries;
- battery electrochemical processes require juxtaposition of multiple phases, including an electrolyte phase that usually obscures the electrode surface or renders it invisible to many experimental probes.

Notwithstanding these difficult problems, Robert Kostecki, Phil Ross, and Tom Devine are using powerful analytic methods, including advanced spectroscopy and microscopy, to characterize materials and cell components and determine the morphological, structural, and compositional changes in electrode materials that accompany cell cycling. Their experimental results have:

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Improved Algorithms Lead to Lab-Scale Combustion Simulations

In spite of the fundamental technological importance of basic combustion processes, our knowledge of them is surprisingly incomplete. Theoretical combustion science is unable to represent realistic flames in all of their complexity, and laboratory measurements are difficult to interpret and often limited in the type of flame and level of detail they can address. Lawrence Berkeley National Laboratory's Computational Research Division (CRD) Center for Computational Sciences and Engineering (CCSE) has teamed with the Environmental Energy Technologies Division (EETD) to build a high-performance computing solution to flame simulation and analysis. This solution allows detailed three-dimensional representation of flame behavior over time, which could not previously be studied using simulation technology. This new capability sets the stage for dramatic progress in combustion science research.

Current investigations are focusing on two primary areas: how turbulence in the fuel stream affects local combustion chemistry and how emissions are formed and released in the product stream. The first detailed simulations of laboratory-scale turbulent premixed flame experiments using CCSE's software have revealed flame chemistry and fluid transport in unprecedented level of detail. CCSE's Mark Day and John Bell, and EETD researchers are working together to validate the simulations with experimental data and to probe the computed results for information not easily obtainable from the experiment in any other way. The work has applications for devices ranging from power generators, to heating systems, water heaters, stove, ovens, and even clothes dryers.

Simulation at this level of detail was impossible just a few years ago. However, algorithmic improvements by DOE-funded applied mathematics groups such as CCSE have slashed computational costs for turbulent flow phenomena by a factor of 10,000. CCSE has implemented its advanced simulation algorithms in state-of-the-art parallel computing hardware to increase the number of variables available for describing a system from hundreds of thousands to more than a billion. These improvements mean that researchers can now simulate real experiments without having to create ad-hoc engineering models for under-resolved physical processes.

CCSE's modeling improvements take advantage of key mathematical characteristics of low-speed flows, which are common to most combustion applications, to eliminate components of the model that are relevant only to high-speed flow scenarios. These components have little effect on low-speed-flow system dynamics, but they drive down simulation efficiency by unnecessarily limiting the maximum numerical time-step size. The integration algorithms are implemented in a set of software tools based on adaptive mesh refinement (AMR), a dynamic grid-based system that automatically allocates computational resources to regions that contain the most interesting detail. The AMR methodology allows researchers to simultaneously incorporate large-scale effects that stabilize the flame and very fine-scale features of the combustion reaction zone itself.

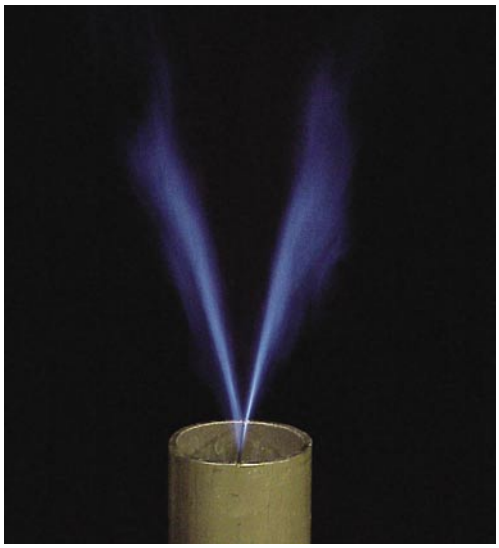

Figure 1. Photograph of rod-stabilized V-flame; blue color indicates location of combustion reaction zone, averaged over duration of camera shutter speed.

The detailed solutions computed by CCSE are being validated with experimental data provided by the EETD Combustion Lab. Preliminary comparisons focus on global characteristics, such as mean flame locations and geometries, as well as statistics of instantaneous flame surface structures. In addition to validating the computed solutions, the research groups probe the vast arrays of data generated by the computations in order to learn more about flame details, such as the localized effects of large and small eddies on the structure of the combustion reaction zone.

For example, the distribution of hydrogen atoms in the thermal field is tightly coupled to key chain-branching reactions required to sustain the combustion process itself. The detailed models accurately represent the transport of hydrogen with respect to the other chemical species in the context of this turbulent flow. CCSE is currently

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using detailed chemistry and transport models containing 20 to 65 chemical species and hundreds of reactions.

Most combustion systems do not operate at low speeds but laboratory experiments where detailed laser diagnostics have been applied are in the low-speed regime. This makes the CCSE method the right one for comparing results with the laboratory experiments.

Example investigations by the group include the simulation and analysis of a three-dimensional V-flame experiment in the EETD Combustion Laboratory (see Figure 1). In this experiment, a thin rod is placed across the exit of a circular nozzle that issues turbulent premixed methane-air fuel vertically into an unconfined region open to the lab. The rod stabilizes a robust V-shaped flame that is highly corrugated and time dependent. Because of the camera's finite shutter speed, the surface of this turbulent flame appears smeared out over a finite flame brush thickness.

Figure 2. These methane images show exceptional agreement between CCSE's simulation (right) and experimental PIV image-data.

In the experiment, the instantaneous location of the flame may be visualized using particle image velocimetry (PIV), in which inert particles are distributed in the unburned gas with a uniform density. The upward-moving gas expands as it passes through the flame; the density of tracer particles shows a corresponding decrease. The location of the abrupt change in particle density, captured in the photograph (Figure 2, left), indicates the instantaneous flame position in a vertical plane through the center of the flame-stabilizing rod.

The photo may be compared to a representative planar slice of the simulated fuel concentration (right) because fuel is consumed at the flame front. The simulation was based on 20 chemical species and 84 fundamental reactions. The computed surface exhibits large-scale wrinkling of the instantaneous flame surface, and, when averaged over time, shows remarkable agreement with the laboratory photos in terms of flame brush thickness, spreading, and growth rates. The still images of the experiment and simulation demonstrate exceptional agreement, both in overall flame shape as well as brush growth characteristics, and fine-scale wrinkling of the flame surface.

The main purpose of using PIV in the experiment is to measure the velocity statistics within the turbulent flame for direct comparison with the simulated results. PIV is a diagnostic method developed for measuring instantaneous velocity distributions within a two-dimensional plane defined by a laser sheet. The system consists of a digital camera, synchronized with a double-pulsed laser beam that is shaped into a thin sheet that dissects the v-flame. The field of view for this PIV system (13 cm by 13 cm in this case), was adjusted to be comparable to the computational domain (12 by 12 by 12 cm). PIV measures the instantaneous velocity vectors by analyzing the spatial displacement of particles shown on a pair of images produced by the double pulsed laser. To obtain velocity statistics, EETD researchers analyzed 448 image pairs. The comparison with simulated results shows that the simulation captures the salient features of the v-flame flow field, including the flame-generated flow deflection, flame-generated acceleration, and buoyancy effects.

CCSE is now working with EETD researchers to develop additional statistical measures of both the simulation and experimental data, which will permit even more detailed quantitative comparisons. The solutions will also be used to track the fate of fuel particles as they wash through the flame and undergo a range of chemical reactions, ultimately contributing to pollutant or unburned hydrocarbon emissions, for this and other interesting experimental configurations. Because processing and understanding the wealth of data generated by these simulations continues to raise questions and because simulation of practical-scale combustion devices remains an immense undertaking, CCSE is constantly building and improving innovative diagnostic techniques that will help shed new light on the complex behavior of turbulent reacting flows.

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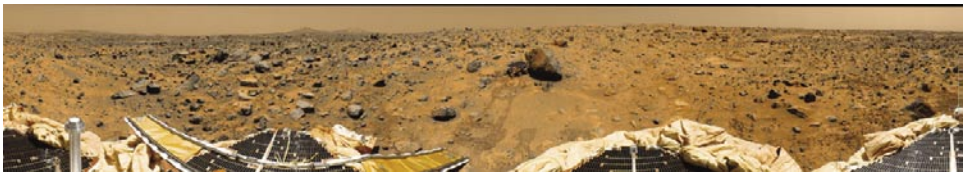

To Get to Mars, Use Wheat

Sometime in the future, in a spacecraft en route to Mars, an astronaut may reach into a container and grab a handful of wheat straw. She'll be holding the key to a sustainable mission, the substance that will convert her incinerated waste into fertilizer for the plants she eats and nitrogen for the air she breathes during her three-year round trip.

Not bad for straw, the inedible portion of wheat used as bedding in horse stables on Earth. Its journey from the farm to Mars comes by way of a team of Lawrence Berkeley National Laboratory (Berkeley Lab) and National Aeronautics and Space Administration (NASA) scientists whose work brings our closest planetary neighbor even closer.

Figure 1. NASA is developing ways to grow wheat using a nutrient solution and artificial sunlight. The crop could feed astronauts destined for Mars, and, thanks to Ted Chang's research, facilitate a continual cycle of food, waste, and nutrients, (Photo: NASA).

"To get to Mars, we need to develop a fully regenerative life-support system," says Ted Chang, a senior scientist in Berkeley Lab's Environmental Energy Technologies Division (EETD) who led the research.

Here's the problem and the opportunity: it's impossible to pack into a tiny spacecraft enough provisions for a whole three-year round trip to Mars, so the crew members will have to grow their own food along the way. This means they'll need enough fertilizer to sustain several harvests.

At the same time, the process of growing and eating food produces waste. The astronauts will have to dispose of not only their own waste but also unused plant fiber. Incinerating these waste products yields reusable compounds like carbon dioxide, water, and minerals, along with noxious pollutants like nitrogen oxides and sulfur dioxide.

This is the making of a short-lived cycle: a dwindling supply of plant fertilizer and a growing pool of pollutants.

But it could also be the making of a sustainable system. Locked in the pollutants are nutrients that can help grow new crops (see Figure 1). Nitrogen oxides can be converted into the fertilizers ammonia and nitrate. They can also be converted into nitrogen to replenish the nitrogen in the spacecraft's air supply. And sulfur dioxide can be converted into sulfate, another fertilizer. Food to waste to nutrients, then back to food; it's a textbook sustainable system.

The trick is stripping nutrients from the pollutants. This process is routine on Earth; one common method uses catalysts with limited life spans, and another relies on spraying an alkaline solution through incinerated waste. However, these methods will not work in space. Short-lived materials will not survive a multi-year mission, and sprays misbehave in a low-gravity environment. "You can't use expendable materials, gravity-dependent processes, or dangerous gases," says Chang. "So we focused on material that is available and can be continuously regenerated."

Chang didn't have to look far. As long as astronauts grow wheat, they'll have a steady supply of straw. Straw that is converted to activated carbon could facilitate a cyclical flow of food, waste, and nutrients.

To test the idea, Chang's team first shreds straw into tiny bits and heats it in an oxygen-

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To Get to Mars, Use Wheat

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free chamber to 600 degrees Celsius. This process converts the cellulose into char, a hydrocarbon product formed during the incomplete burning of organic material. Next, the char is activated by heating in the presence of carbon dioxide or water, which breaks the char's carbon-carbon bonds and increases its surface area and porosity. The broken carbon bonds also create unpaired electrons that are ready to bind with new compounds.

Figure 2. Illustration of the Mars rover, (Photo: NASA).

The activated carbon is placed inside a steel tube and exposed to a gaseous stream of incinerated waste and its pollutants. With the aid of oxygen, nitrogen oxides are grabbed by the carbon column's unpaired electrons and adsorbed onto its pores. A final step, in which the column is heated, regenerates the activated carbon and converts the adsorbed pollutants into nitrogen gas. Alternative steps yield other useful compounds: exposing the column to water produces nitrate, and, if the column has adsorbed sulfur dioxide as well as nitrogen oxides, exposing the column to water produces ammonia. To replenish the small portion of activated carbon lost in the final heating step, the astronauts can simply harvest more wheat straw.

But is this process sustainable month after month? Early calculations are optimistic. A six-person crew would eat 1.5 kilograms of wheat per day, which could yield 203 kilograms of wheat-straw-derived activated carbon each year—enough to supply the crew's needs.

"Waste has nutrients that shouldn't be thrown away, and in fact could help sustain a mission for its entire duration," says Chang. "Our method could allow astronauts to reuse valuable resources."

To further evaluate the Berkeley Lab system, scientists at the NASA Ames Research Center at Moffet Field, California, are currently conducting large-scale tests. The research appears in the September/October 2003 issue of *Energy & Fuels*.

—Dan Krotz

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- provided new insight into unwanted side reactions that take place along with the main electrochemical reactions in rechargeable batteries,
- revealed unexpected physical changes that take place on electrode surfaces as BATT baseline cells degrade, and
- shed light on corrosion processes on the metal foils used as substrates for cathodes in lithium-ion batteries.

Sophisticated mathematical modeling is required to understand the complex phenomena in batteries. As noted previously, mathematical models are being used to optimize the design of BATT Program baseline cells (see Figure 3). Comprehensive stability analysis is also being designed to determine the conditions under which lithium-dendrite growth can be inhibited or eliminated by mechanical means. This type of modeling can be extremely cost effective because it relies on rapid computer simulations rather than slow and expensive experimental approaches to address fundamental problems.

Outlook

The BATT Program has changed immensely during the past several years, incorporating the latest experimental and modeling tools to address fundamental battery problems and keeping pace with the rapid introduction of the lithium-ion battery. The BATT Program has played and will continue to play a leading role in battery research and is currently poised to help introduce the lithium-ion "superbattery" to the HEV market. The ongoing work of BATT researchers is essential to a future with cleaner vehicles and less dependence on petroleum.

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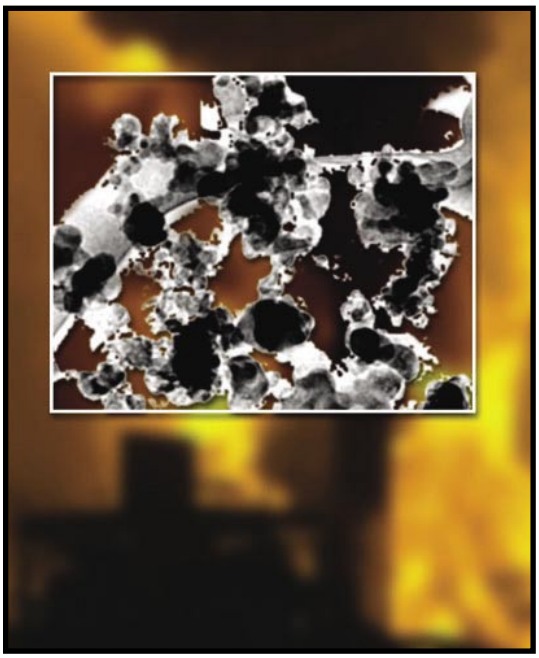
CARBONACEOUS AEROSOLS and CLIMATE CHANGE:



How Researchers Proved Black Carbon is a Significant Force in the Atmosphere

They can absorb light or scatter it. They are present in the atmosphere because of the incomplete combustion of fossil fuels. Now they are thought to have a significant effect on global warming. But until just 10 or 15 years ago, the scientific community did not accept that carbonaceous aerosol particles were common in the atmosphere. That they accept this idea now is thanks to the work of a research group, led by Tihomir Novakov at Lawrence Berkeley National Laboratory (Berkeley Lab), which has been studying these particles since the 1970s.

Carbon aerosol (CA) particles are composed of light-scattering organic carbon (OC) and light-absorbing black carbon (BC). These particles are important to the atmosphere because they can block solar radiation and scatter visible light and because they are as common as sulfates, which are well-known particles found in the atmosphere as a result of the burning of fossil fuels.

Both carbon and sulfate particles can affect climate in ways that either increase or reduce the effects of global warming. Sulfate and carbon (particularly OC) aerosols scatter light back to space, which reduces the warming caused by greenhouse gases. BC alone has a dual effect: it absorbing sunlight, which heats the atmosphere, but also blocks light from reaching Earth's surface, which has a cooling effect. An additional, indirect effect of aerosols is to increase the reflectivity of clouds (i.e., make cloud surfaces "shinier"). This change in reflectivity reflects the sun's heat back into space, which reduces global warming. Climatologists are now trying to understand the sum of all of these effects on global climate change.

A Voice in the Desert

Novakov, who has been called the "godfather of black carbon studies" by eminent National Aeronautics and Space Administration climate researcher James Hansen, heads the atmospheric aerosol research group in what is now Berkeley Lab's Environmental Energy Technologies Division (EETD). This research group began studying carbon aerosols during the 1970s; their work was one of the division's earliest research areas. In 1974, Novakov, with aerosol research group member S.G. Chang as well as A.B. Harker, published a paper in Science claiming that carbon constitutes 50 percent of the total particulate concentration in urban atmospheres and that as much as 80 percent of particulate carbon is in the form of soot or BC.

"We were a voice in the desert," says Novakov. "It was an unconventional view, and it was long time before the scientific community agreed with us even though it made sense to some." The conventional view at the time was that sulfate and OC aerosols were produced primarily by photochemical smog reactions in the atmosphere. Many researchers in the United States thought that BC was insignificant in the atmosphere, no longer present since the Industrial Revolution had ended and the number of coal fires used to generate heat in homes and factories was dwindling. But evidence from Novakov's group would change this thinking.

"What [Novakov] did," says Lara Gundel, a scientist in his group during the 1970s, "is ask 'why air pollution is black?' If air pollution was formed photochemically, then what was the black

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component?" Gundel, who continues to research organic and particle air pollution at Berkeley Lab today, adds, "in the 1970s, the air pollution community was all about smog, and how ozone contributes to its formation. There was not much interest in these particles."

Figure 1. The measurement station (small building, right) at Point Reyes lighthouse, Northern California.

To prove their assertion, Novakov, Ray Dod, Dick Schmidt, and other colleagues began to develop new measurement approaches. A 1977 paper, for example, reviews the use of electron spectroscopy for chemical analysis (ESCA) methods for making the first attempt to characterize the chemical components of particulate carbon. By comparing the spectrum of carbon at room temperature with that of carbon that had been heated (heating drives off volatile organic carbon), they determined that most of the black carbon in air samples was soot: inorganic, not organic carbon. (ESCA identifies chemical composition by measuring the spectra of electrons under x-ray bombardment).

Figure 2. Sampling location at El Yunque Peak, Puerto Rico. The instrument measures the light reflected from cloud drops.

Figure 3. Interior of the hut on El Yunque where instruments measure the cloud drop concentrations and aerosols that serve as cloud drop formation (condensation) nuclei.

On a Solid Footing

By the 1980s, the scientific community had begun to take the group's "black carbon hypothesis" more seriously. General Motors sponsored a symposium in 1980 in Warren, Michigan (Particulate Carbon: Atmospheric Life Cycle) during which Novakov delivered an address on soot in the atmosphere. He first used the term "black carbon" in this paper as he reported on his group's work to quantify soot in various U.S. cities' atmospheres and on the increasing weight of evidence that black carbon was a substantial part of the atmosphere's particle burden.

During the 1980s, Novakov frequently described black carbon as "produced solely by the incomplete combustion of fuels," asserting black carbon particles were as important in contributing to both local and regional air pollution (e.g., the Arctic haze) as other pollutants like sulfates. "I tried to emphasize that they were interesting because they were a measure of inefficient combustion," he says.

However, the group still needed better measurements of BC particles and their persistence in the atmosphere over time. During the early 80s, Hal Rosen and Tony Hansen, who were then physicists in Novakov's group began applying optical methods for characterizing and measuring BC. Rosen used Raman spectroscopy to unambiguously demonstrate that BC is composed of graphitic carbon. Rosen put together a simple device that measured the absorption of light by black carbon deposited on a filter with air passing through it.

"I deduced that the rate at which the filter became black with carbon was proportional to the amount of carbon in the air," says Hansen. From this initial model, Hansen, Rosen, Novakov, and others developed a real-time measurement device, which they called an "aethalometer" and described the device in a 1984 paper. ("Aethalon" is a Greek word meaning "to blacken with soot.") Gundel's work was essential in making the aethalometer a quantitative device for measuring BC concentrations.

Hansen built a number of aethalometers. One of the first was used for a study, which began in 1983, of haze in the Arctic atmosphere. Hansen continued to work for Berkeley Lab and also began building aethalometers for researchers around the world as a private consultant. One of Hansen's instruments, built for the Canadian Arctic's Alert research station, is still functioning and has been measuring BC particles for 15 years.

Indications of Long-Distance Transport

Once reliable measuring devices were available to detect BC and sampling and analysis techniques permitted scientists to measure the chemical content of atmospheric particles, it became possible to determine how widely BC particles were spreading across the globe from their origins in cities and industrial areas. During

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the late 1970s and early 1980s Novakov, Rosen, other members of their group, and investigators elsewhere sampled the Arctic atmosphere to determine whether BC particles could be found in what was thought to be a pristine environment. Papers published during these years reveal substantial concentrations of soot found throughout the Arctic. In one study, Novakov and colleagues found BC particles in the Alaskan, Canadian, and Norwegian Arctic regions.

In a 1983 paper, Rosen and Novakov wrote, "These results show that the large concentrations of particles found at the Barrow Alaska site are not a local phenomenon but are characteristic of ground-level stations across the western Arctic." They speculated that "these highly absorbing particles" could have a considerable effect on the Arctic radiation balance and climate. Novakov says he concluded at the time, "If it was in the Arctic, it must be everywhere."

Finding BC particles, which are anthropogenic (produced by human activity, i.e., the incomplete combustion of fossil fuels), in the Arctic signified that human activities were having an effect in the farthest reaches of the globe. "It was obvious that this was evidence of long-range transport," says Hansen.

Of even greater concern was that the heat-absorbing properties of BC could have a warming effect in the Arctic. The work created a stir in the scientific community, which now had direct proof of the long-range transport of pollutants and also now had another question to answer: what effect were BC particles having on Earth's climate?

In 1987, Novakov worked with a group of scientists at New Mexico State University to measure the graphitic content of carbon taken from snow samples in New Mexico, Texas, Antarctica, and Greenland. Black carbon's presence in all of these samples, and its detection in other studies, showed that it had settled out from the atmosphere all over the world. (In a recent paper in the Proceedings of the National Academy of Sciences, James Hansen of NASA's Goddard Institute for Space Sciences, argues that black carbon on the surface of Earth's ice caps absorbs heat from the sun, accelerating the melting of these caps as well as increasing global warming.)

The Effect of Black and Organic Carbon on Cloud Formation

The emergence the data gathered by Novakov and his fellow researchers brought increasing attention to black carbon particles from a scientific community that, by the late 1980s and early 90s, had accepted the presence of BC in the atmosphere as proven. Researchers began to focus on understanding the relative contributions of BC's absorption and scattering of light, which increases global warming, and its intensification of the heat-reflection of clouds, and blocking sunlight, which cools the atmosphere.

Novakov turned his attention to the effect of BC on cloud formation and asked whether the other component of carbon aerosols, the OC particles, were present in the atmosphere in high enough concentrations to affect cloud formation. Again, Novakov's hypothesis went against the conventional wisdom, which was that sulfates, not carbon, were the major force in cloud formation.

In a 1993 paper, Novakov, along with J. Penner of Lawrence Livermore National Laboratory, demonstrated that organic carbon is as effective a nucleus as sulfate particles for the formation of cloud droplets. This finding showed that sulfates were not the only significant man-made source of cloud nucleation in the atmosphere, as had been previously thought.

Novakov published two papers in 1997, with Peter Hobbs and other colleagues at the University of Washington, reporting measurements of aerosols on the east coast of the U.S. Both studies demonstrated that, in some locations, carbon aerosols contribute more than sulfates to the extinction of solar radiation in the atmosphere. "I think this work was a turning point," says Novakov. These papers were cited extensively, and the scientific community accepted another Novakov hypothesis about carbon particles: that these particles compete with sulfate in creating climate change. The scientific community began focusing attention on what BC and OC particles were doing to climate.

Figure 4: Left to right: Dick Schmidt, Tihomir Novakov and Henry Benner with sampling equipment they built, used in European studies.

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Carbonaceous Aerosols and Climate Change

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Figure 5. Dick Schmidt inspects carbon aerosol sampling equipment

"Novakov's persistence, clarity of thought, and ability to be a maverick has led to real progress in this field," says Gundel. "…He asked interesting questions without being part of the mainstream. The mainstream eventually caught up to him." Novakov's impact has been felt not only in his own research but also in his role in organizing seven international conferences on carbon particles in the atmosphere, starting in 1978. The eighth conference will take place in September 2004 in Vienna, Austria.

A Summation, and the Future

One legacy of Novakov's group comes from the work of its alumni. Tony Hansen has continued to build aethalometers and to work in the Lab's Engineering Division, developing state-of-the-art instrumentation for a wide variety of projects from every scientific discipline. S. G. Chang continues to work in EETD, developing methods of improving air pollution control processes and technologies to reduce power plant emissions. Hal Rosen now works at IBM. Lara Gundel researches new methods to accurately measure semi-volatile and particulate organic pollutants in ambient air and combustion sources. She won an R&D 100 award in 2000 for developing a fine sorbent coating, used in air-sampling devices called diffusion denuders, to improve the accuracy of sampling of airborne particles. Ray Dod is retired but still works with Gundel on organic pollutant studies. Dick Schmidt continues to contribute to the design and development of aerosol instrumentation at Berkeley Lab. Others from the early days of Novakov's group (in 1978 there were 18 staff scientists and research associates in the group) either still work at the Lab or did so until they retired.

Carbon aerosol research continues at Berkeley Lab with the involvement of new researchers. Recent papers from the Novakov group focus on the history of BC concentrations in the atmosphere to determine the contribution that BC particles have made to climate change over time. The next article in this series will address this research.

—Allan Chen

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Research Highlights

Agenda 21 Energy-Efficient Office Building Now Open

Environmental Energy Technologies Division (EETD) scientist Joe Huang recently visited the Agenda 21 Energy-Efficient office building in Beijing, China that he and other scientists helped to design. The building was officially dedicated in January when U.S Department of Energy Secretary Spencer Abraham paid a visit.

The new building houses the Technology Development Office of China's Ministry of Science and Technology and has become a showpiece of Chinese energy-efficient buildings, receiving much publicity and media attention since it opened.

Huang was involved with the design of the building as early as 1999 when he performed a feasibility study for the lighting system, which has both occupancy and daylight sensors. Also incorporated into the design were custom-made light shelves, which he had recommended.

During the dedication, Secretary Abraham lauded the building as a demonstration of how the U.S. and China can work together to promote clean energy solutions.

Scientist Recognized for Advancing Wind Energy Markets

In late March, at its first worldwide event, the American Wind Energy Association (AWEA) recognized a diverse group of business leaders, advocates, and policymakers as the most influential voices in the development of the wind power industry. Among those recognized with a special achievement award was Environmental Energy Technologies Division (EETD) scientist Ryan Wiser. Wiser, of the Energy Analysis Department's Energy Policy Analysis Group, was honored for "excellence in research, analysis, and clear documentation of renewable energy policy and market drivers." Wiser has published a strong body of work related to wind and other renewable energy sources, including noteworthy research on renewable energy policy and economics.

"The breadth and scope of the wind industry's impact on the U.S. and, indeed, the global economy is reflected in this year's distinguished award winners," said AWEA Executive Director Randall Swisher. "The success in developing America's growing wind energy market is due in large part to the leadership of these organizations and individuals who demonstrate their commitment to renewable energy every day in their actions."

Wiser says, "I greatly appreciate this award from the AWEA. Wind power is on the cusp of greater market penetration in the U.S. and worldwide, and I have valued being a part of this process. I'd like to thank AWEA for this distinction, as well as my stellar staff for helping me achieve the award."

EETD Scientist Receives Fellowship

Environmental Energy Technologies Division (EETD) Staff Scientist Jonathan Koomey, now a visiting professor with a joint appointment in the Engineering and Earth Sciences departments at Stanford University, is one of 20 recipients of an Aldo Leopold Leadership Fellowship for 2004.

Aldo Leopold Leadership Fellowships offer scientists intensive communications and leadership training to help them convey scientific information effectively to non-scientific audiences, especially policymakers, the media, business leaders, and the public. Fellows are selected through a competitive application process and must have outstanding scientific qualifications, demonstrated leadership ability, and a strong interest in communicating science to audiences outside the traditional academic arena.

The program is named for Aldo Leopold, a renowned environmental scientist who communicated his knowledge simply and eloquently. His writings, including his 1949 book, A Sand County Almanac, are credited with infusing the emerging conservation movement with good science and a stewardship ethic.

Jon Koomey comments, "I'm excited to be a Leopold Leadership Fellow for 2004. This award will help me hone my skills in explaining complex technical issues to policymakers and the public."

Research Highlights

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EETD Scientist Publishes Important Dosimetry Treatise

An important book on the effects of exposure to aerosols has been co-authored by Lev Ruzer, a visiting scientist in EETD. *Aerosols Handbook: Measurement, Dosimetry, and Health Effects* will be published by CRC Press (http://www.crcpress.com) in October of this year. The handbook is a collaborative effort of some 30 researchers, among them EETD researchers Lara Gundel, Richard Sextro, Phillip Price, and Michael Apte.

Ruzer studied particle deposition in the lungs of uranium miners when he worked in Tadjikistan, which was then a part of the Soviet Union. That body of work was not published but is incorporated and updated in this upcoming volume. Additional studies with aerosols in outdoor and indoor exposures, industrial situations, pharmaceuticals, and bioaerosols have been included. Important overlooked areas, according to Ruzer, are dosimetry methods and the health effects of aerosol deposition. Dosimetry methods for uranium miners were first developed by Ruzer during the 1970s. He characterized that work as "the most difficult project of my professional life because of the secrecy which surrounded the research. Now many will be able to understand how deposition takes place."

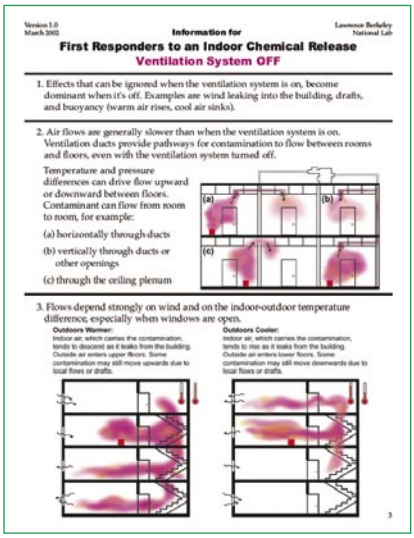
Recognition for Efforts to Protect Buildings from Chemical and Biological Weapon Attacks

A team from EETD has won a Federal Lab Consortium (FLC) Award for Excellence in Technology Transfer in 2004.

The winning entry is *"Minimizing Casualties from a Chem/Bio Attack: Preparation, Training, and Response Resources."*

The team members includes the following from Lawrence Berkeley National Lab: Ashok Gadgil, Phil Price, Tracy Thatcher, Michael Sohn, David Lorenzetti, Rengie Chan, Emily Wood, Woody Delp, Sondra Jarvis, Richard Sextro, Elizabeth Finlayson, Buvana Jayaraman, Sheng-chieh Chang, and Seungbae Hong. Other members of the team include William Nazaroff of the Department of Civil and Environmental Engineering, University of California, Berkeley; Gayle Sugiyama, Lawrence Livermore National Laboratory; and Susanna Gordon and Donna Edwards of Sandia National Laboratories, Systems Research Department.

In late 2001, terrorists used anthrax to kill several people, disrupt mail deliveries, and render congressional office buildings in Washington D.C. uninhabitable. The buildings were eventually reoccupied at a cost of more than $150 million and after enormous disruption to their occupants. These relatively limited attacks had huge consequences; a major chemical or biological attack could be much more severe.



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Information for

Lawrence Berkeley National Lab

First Responders to an Indoor Chemical Release

Ventilation System OFF

1. Effects that can be ignored when the ventilation system is on, become dominant when it's off. Examples are wind leaking into the building, drafts, and buoyancy (warm air rises, cool air sinks).

2. Air flows are generally slower than when the ventilation system is on. Ventilation ducts provide pathways for contamination to flow between rooms and floors, even with the ventilation system turned off.

Temperature and pressure differences can drive flow upward or downward between floors. Contaminant can flow from room to room, for example:

(a) horizontally through ducts

(b) vertically through ducts or other openings

(c) through the ceiling plenum

3. Flows depend strongly on wind and on the indoor-outdoor temperature difference, especially when windows are open.

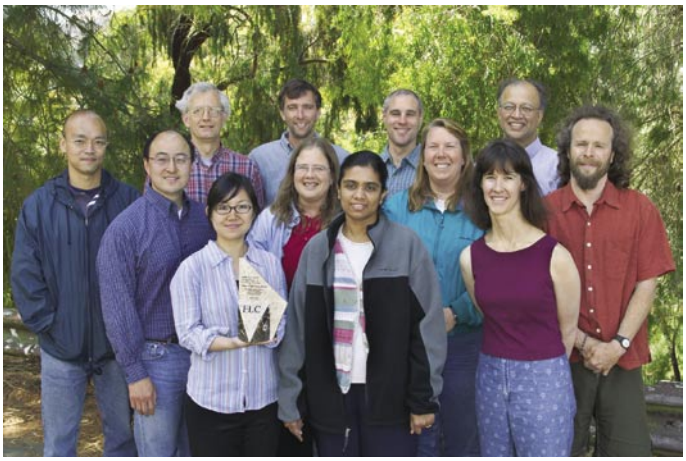
Outdoors Warmer:
Indoor air, which carries the contamination, tends to descend as it leaks from the building. Outside air enters upper floors. Some contamination may still move upwards due to local flows or drafts.

Outdoors Cooler:
Indoor air, which carries the contamination, tends to rise as it leaks from the building. Outside air enters lower floors. Some contamination may still move downwards due to local flows or drafts.

Securebuildings.lbl.gov provides materials to train first responders in the event of a chemical or biological attack on a building.

Even before the anthrax releases, scientists in EETD's Indoor Environment Department had been conducting research on reducing the effects of a chemical or biological attack. This research builds on a long tradition of work on building airflows, filtration effectiveness, and air quality issues. The attacks prompted scientists to ask, "Is there anything we can contribute right now?"

The answer was "yes." They identified several target groups that could benefit from increased knowledge, including building operators, who are responsible for the maintenance, and operation of building ventilation systems; managers of unique, high-value buildings such as airports; emergency planners and incident commanders who have to decide what areas of a city to evacuate and where to send response teams; and "first responders," the firefighters and police officers who are the first trained people on the scene of an attack.



Left to right, front row: Rengie Chan, Buvana Jayaraman, Sondra Jarvis; center, Michael Sohn, Elizabeth Finlayson, Tracy Thatcher, Woody Delp; rear, Sheng-chieh Chang, Rich Sextro, Phip Price, David Lorenzetti, Ashok Gadgil.

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Research Highlights

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The EETD team developed advice for each of these groups through the Secure Buildings website, which has had thousands of visitors since early 2002. EETD researchers worked with colleagues at Sandia National Laboratories to make recommendations to airport managers on preparation, training, and event response. They also worked with the National Atmospheric Release Advisory Center at Lawrence Livermore National Laboratory to predict indoor toxic concentrations from a Bhopal-type accidental chemical release, an important addition to the suite of outdoor prediction tools already available, because in industrialized nations people spend the majority of their time indoors.

Finally, the team created First Responder training materials for the California Peace Officers Standards and Training Agency, which has used the materials to train police officers in much of the U.S.

These efforts have improved the readiness and safety of the nation's police officers, the security of the nation's buildings and their inhabitants, the effectiveness of local emergency response, and the safety of the U.S. air transportation network.

For more information, see http://securebuildings.lbl.gov

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With more than 4,000 employees, Berkeley Lab's total annual budget of nearly $400 million supports a wide range of unclassified research activities in the biological, physical, computational, materials, chemical, energy, and environmental sciences. The Laboratory's role is to serve the nation and its scientific, educational, and business communities through research performed in its unique facilities, to train future scientists and engineers, and to create productive ties to industry. As a testimony to its success, Berkeley Lab has had nine Nobel laureates. EETD is one of 13 scientific divisions at Berkeley Lab, with a staff of 400 and a budget of $40 million.

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