Fulfilling the Promise of the Materials Genome Initiative with High-Throughput Experimentation

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Executive Summary

The Materials Genome Initiative (MGI), a national effort to accelerate and lower the cost of advanced materials technologies, has made significant progress in computational simulation and modeling of new functional materials. To build on this progress, a large amount of experimental data validating these predictions and informing more sophisticated models will be required. High-throughput experimentation (HTE) generates large volumes of experimental data using combinatorial materials synthesis and rapid measurement techniques, making it an ideal partner to bring the MGI vision to fruition. A workshop entitled "Combinatorial Approaches to Functional Materials" was held in San Francisco on May 5-6, 2014 to explore opportunities for HTE to advance the MGI. This paper summarizes the findings of the workshop, and proposes a national effort in accelerated HTE materials innovation to address challenges of national and global importance.

Introduction

The discovery and commercialization of advanced materials is crucial to solving major challenges in security, environment, and economic growth. In 2011, President Obama announced the Materials Genome Initiative (MGI), a national effort to accelerate and lower the cost of advanced materials technology development. The MGI approach requires contributions in three critical areas: computational tools, experimental tools and digital data. In three years, the MGI has made significant progress in predicting new functional materials through computational simulation and modeling of materials properties. To build on this progress, large amounts of experimental data validating these predictions and informing more sophisticated models will be required. High-throughput experimentation (HTE) methodologies, which utilize materials "libraries" to rapidly determine physical and chemical properties, are uniquely suited to provide the experimental counterpart to computational efforts and bring the vision of the MGI to fruition.

A workshop entitled "Combinatorial Approaches to Functional Materials" was held in San Francisco on May 5-6, 2014[1] to explore opportunities for HTE to advance the MGI. Attended by 90 experts from government, industry, and academia, the workshop was co-organized by Applied Materials and the University of South Carolina with support from the National Science Foundation (NSF), the National Institute of Standards and Technology (NIST) and the White House Office of Science and Technology Policy (OSTP). The workshop assessed current HTE capabilities, outlined challenges, and identified opportunities for significant impact.

This report summarizes the findings of the workshop, and proposes a national effort in accelerated materials innovation through HTE to address challenges of national and global importance. Advances in new materials for the energy, electronics, chemical, aerospace, and defense sectors will provide technology solutions, drive economic growth, and enhance national security. These advances will additionally bolster the competitiveness of the United States in the 21st century and create a highly skilled national manufacturing workforce.

major challenges	materials solutions
national security	low-weight, high-strength transportation materials extended life batteries radiation detecting materials solar powered military equipment high sensitivity magnetometer materials
dependence on critical raw materials	reliably available catalysts and permanent magnets high-temperature turbines
climate change	high-efficiency, low-cost solar cells strong, increased-efficiency wind turbines materials for CO_2 absorption, capture and chemical conversion fuel cell and solar fuels catalysts materials for CH_4 storage, conversion and utilization chemical and biological sensors
electronics demand	smaller, higher-performance transistors atomic control over device interfaces
economic growth	cost-effective materials for increased U.S. production and manufacturing new hardware and materials-based technologies

Table 1. Timely discovery and commercialization of advanced material technologies are crucial to addressing major national and global challenges.

Why Now?

Today's pressing national challenges—manufacturing competition, dependence on critical raw materials, and climate change, to name a few—underscore the urgent need for advanced materials solutions (**Table 1**). Advances in HTE methodologies have been successfully demonstrated in every class of technologically important materials[2-10] and may be used to discover and deploy new materials at unprecedented speed and low cost. However, a full realization of the MGI vision will require a carefully planned integration of experiments, computation, and theory; open access to high quality digital data and materials informatics tools; and a well-equipped workforce. A coherent and efficient mobilization of the HTE community will create high-impact solutions for the pressing needs ahead.

Workshop Results

Major conclusions from the recent workshop were that:

- Critical technologies such as energy production and utilization, microelectronics, and catalysis await immediate materials solutions through the discovery and development of higher-performance photovoltaic, thermoelectric, energy storage, fuel cell, semiconductor and catalytic materials
- Existing HTE programs, while producing excellent core technologies, lack efficient coordination, resulting in underdeveloped opportunities and capabilities in data collection, curation and analysis
- The widespread utilization of HTE methodologies is required to complement, inspire and validate computational MGI efforts
- The HTE methodology is uniquely suited to rapidly generate the large volumes of high-quality materials data required to populate materials properties databases
- Standards for library synthesis, characterization, and data curation (e.g., library architectures, data formats, and informatics) are crucial for effective and widespread use of HTE investments

Key recommendations from the workshop were to:

- Enable broad access to HTE methods and data through the creation of multiple user centers with library synthesis, metrology and informatics capabilities
- Establish public/private collaborations that accelerate commercialization in key advanced materials arenas including electronic materials, catalysts, and energy-related materials
- Develop new library design and characterization techniques that extend the scope of HTE research beyond composition space to include the study of surfaces, interfaces, processes and integrated devices; focus efforts on specific properties relevant to end-use applications of new materials; and create libraries that include parameters such as processing variables, interface properties (transport, electronic, phonon, and grain boundary effects), and performance measures (e.g., device reliability)

Opportunities

There are many areas that can immediately benefit from the proposed HTE approach. For example, low cost solar cells made from earth-abundant materials will play an important role in developing sources for clean, renewable energy. The development of new solar cells requires a combination of up to six property-matched materials to be integrated into a complex multilayered device structure with specific and demanding performance metrics. HTE methods can accelerate the search for novel light absorbing materials free of expensive indium or toxic cadmium, and rapidly optimize high performing materials combinations.

Another energy application is the significant opportunity for waste heat recovery using thermoelectric devices. Recent studies have shown that for the U.S. alone, annual potential for electrical energy recovery from waste heat could be in the multi-terawatt range[11]. Energy storage materials such as in Li ion batteries represent another opportunity for the HTE approach. Today each electrode/electrolyte combination is tested individually through a set of controlled electrochemical experiments. This approach is tedious and slow due to the large number of electrode materials (e.g., mixed metal oxides, phosphates, graphite) and electrolytes (solvent blends, co-solvents and additives) involved. HTE can rapidly screen for charge transfer kinetics and battery stability in order to determine optimum combinations of electrode and electrolyte.

In the last 20 years, the number of elements (and therefore new materials) used in microelectronics manufacturing has jumped from 12 to over 50 (**Figure 1**). As device dimensions approach a few atomic layers, electronic wave functions and materials properties begin to be dominated by surfaces and interfaces and can no longer be estimated from bulk behavior. The ability to understand and build lower power switches such as tunneling field-effect transistors (TFETs) and interconnects with lower resistance is fundamentally linked to an accurate understanding of atomic-scale interfaces. A wealth of detailed experimental data will fuel the design of complex three-dimensional structures on the atomic scale.

Growth in the nascent "Internet of Things" will be catalyzed through sensor development, an area that requires advanced electronic technologies including flexible and post-CMOS (e.g. graphene) devices. The variable performance of sensors with changing environmental conditions like humidity, temperature, and pressure is a significant concern. Again, HTE can be used to rapidly characterize the variation in sensor response and validate optimal sensor designs.

Catalysis is at the core of future energy technologies including syngas production via biomass conversion, artificial photosynthesis, water splitting, heterogeneous catalysis of methane activation, fuel cell electrocatalysis and solar fuels reactions. For many of these applications, state-of-the-art catalysts involve high loadings of precious metals; the development of reliably available or reduced platinum-group metal catalysts will favorably shift the economic viability and environmental impact of these applications. Catalyst development is an ideal HTE target since heterogeneous catalytic activity is a complex function of the composition, surface and bulk structure, and interactions between the catalyst and the underlying support. For catalysis and many other high technology materials applications, there is an urgent need to find substitutes for critical and expensive materials, or materials whose supply chain is unstable. Rare earth elements comprise an important class of critical materials, with other examples including rhenium for jet engine components, indium for solar cells and display technologies, and platinum for fuel cell electrodes. HTE methodologies are an invaluable tool for materials substitution research.

Currently, the lack of high quality data presents a barrier to the realization of materials solutions for critical technologies. HTE can lower this barrier by populating databases through the generation of large-volume, high-quality experimental data. A national database with comprehensive computational and experimental data on electronic materials, for example, may enable in silico designs and protocols for novel integrated device fabrication. Such databases would be a significant resource for materials engineers in industry, academia, and government, and should be established.

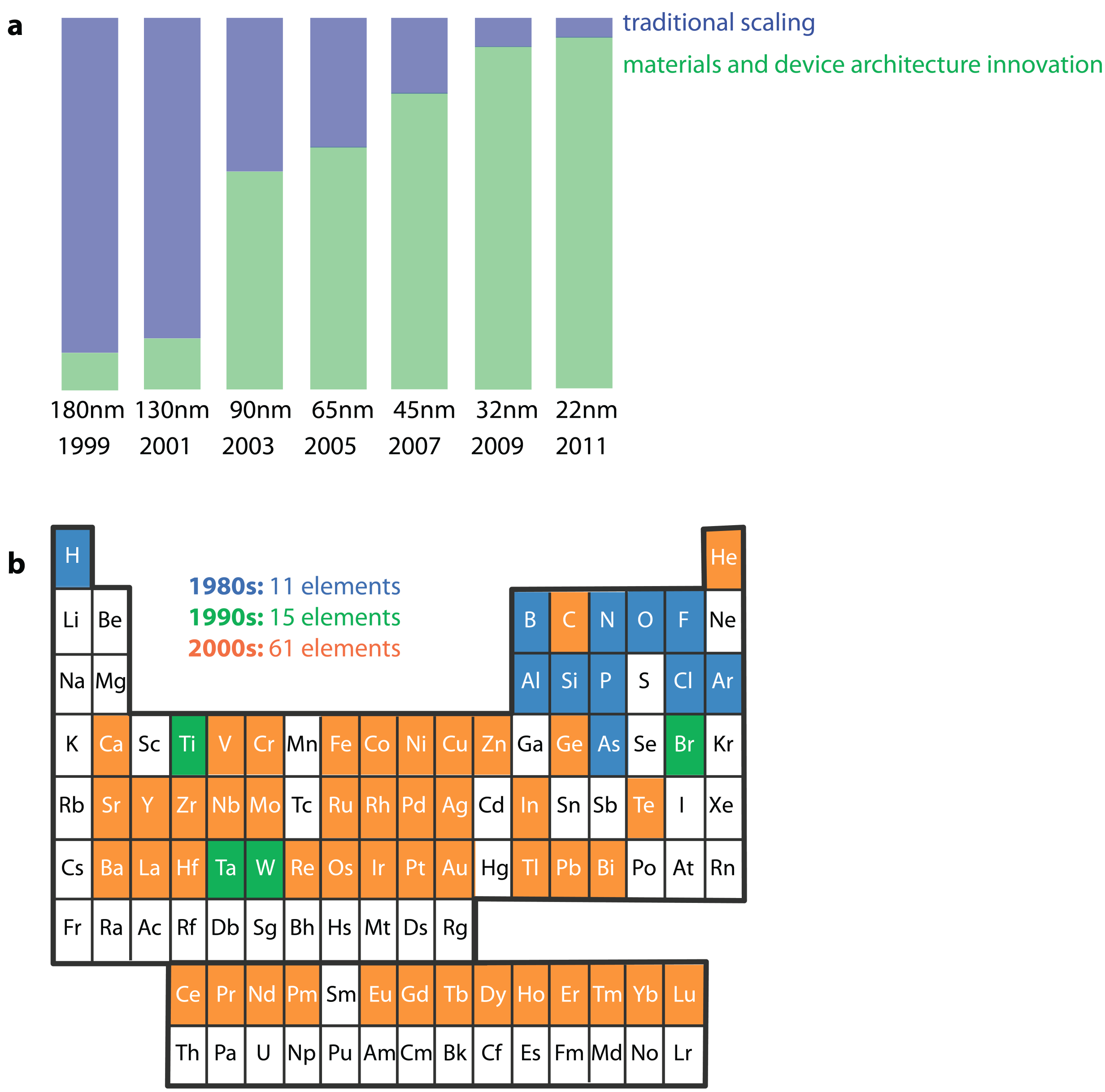

Figure 1. Advances in materials innovation are critical to continued growth of the semiconductor industry. (a) The fraction of R&D spending by the semiconductor industry associated with traditional "scaling" vs advanced materials development for each generation of Moore's Law, highlighting the critical nature of accelerating advanced materials development for commercialization. **(b)** Periodic Table of the Elements, showing the atomic materials used for semiconductor manufacturing from the 1980s to today. The rapid increase in the breadth of advanced materials must be considered when developing new semiconductor devices.

Challenges for HTE

The HTE approach consists of three major steps (**Figure 2**): 1) hypothesis-driven design and synthesis of a "library" sample that contains variations of the materials parameter of interest (typically composition); 2) rapid, local, and automated interrogation of the library for the properties of interest; and 3) acquisition, curation, analysis and mining of the resultant data. Each step presents challenges that must be overcome before HTE methods can be reliably deployed.

Library synthesis and metrology tools are often expensive, not readily available commercially, and not accessible to most scientists. They require dedicated staff to operate, calibrate and maintain them. HTE characterization tools for materials structure, e.g., x-ray diffractometers, are available at synchrotron beam lines, but HTE methods to characterize surfaces, interfaces or chemical bonding have not yet been developed. Additionally, for HTE to effectively address MGI goals, the data must be curated, analyzed, and mined to rapidly obtain new knowledge. This presents an increasing challenge not only in materials engineering but also in a number of technical disciplines due to growing data generation and storage.

As currently practiced, none of the three steps of the HTE approach are standardized, an issue that must be resolved to establish an efficient national effort. Standard library sizes, measurement layouts, and data formatting will facilitate sharing of HTE tools and comparison of data in disparate locations. HTE progress can be further accelerated through an expansion of materials property databases far exceeding the archived materials data available today. Online databases of materials properties, all in digital formats for exchange, access, and preservation of HTE datasets, will ensure searchability and interoperability. Standard library film samples for selected properties (e.g., phase distribution and dielectric constant) can be made available with certified measurement values under specified experimental conditions (e.g. temperature) to calibrate disparate HTE metrology tools. Analysis software must be standardized and open access granted. These practices will increase the level of confidence in HTE data generated among individual laboratories.

Currently, HTE libraries and measurement tools are primarily designed with the goal of materials discovery and optimization, resulting in composition as the typical variable parameter. However, the properties of a material are governed by structure as well as composition, and HTE methods to sample such structural landscapes must be devised[9-10]. Furthermore, thin film composition libraries are not traditionally deposited using the processing tools and conditions required to produce the intended device. Library design must evolve so that the libraries increasingly represent the actual materials and processing conditions that the selected materials will be exposed to during device production. Solar cells, fuel cells, and transistors, for example, all require the integration of multiple materials with different functionalities. These devices all contain numerous interfaces, such as the p-n junction in photovoltaics, or catalytic surfaces at the solid-water interface in photoelectric water splitting. The properties of these interfaces are the defining functional property of the devices enabled by these materials. Therefore HTE techniques should be additionally applied to processing parameters, interface engineering, and performance of multi-layer stacks/devices. To make this evolution possible, it is necessary to develop HTE metrologies that measure interface properties (e.g., electronic, magnetoelectric, phononic), grain boundary effects on device performance, and reliability.

Finally, while HTE synthesis tools have been optimized for the production of thin films, many new bulk materials are in demand, including lightweight structural materials for transportation and rare earth-free magnetic materials in wind turbines. There is, therefore, additional need for the development of HTE bulk materials synthesis techniques.

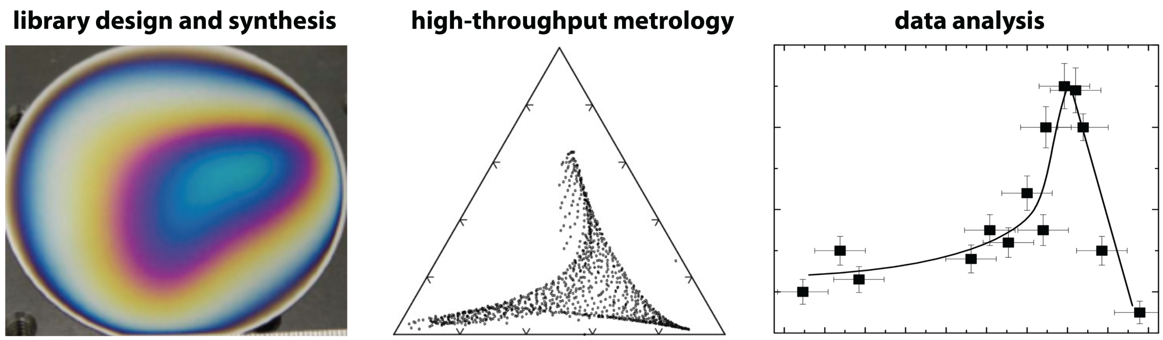

Figure 2. The three components of high-throughput experimentation (HTE) are used to rapidly characterize and create new insights on a materials landscape. HTE methodologies enable the creation of large amounts of experimental materials data, providing the opportunity for new insights and a greater understanding of the effects of composition and processing methods on numerous materials properties. Future metrology tools aim to additionally investigate properties of materials interfaces and device performance.

Required Infrastructure for HTE, and Present Gaps

Over the past decade, there have been several pioneering HTE efforts. However, many of the critical components of a highly productive HTE platform, such as next-generation metrologies, effective data analytics, and robust communication between computational and experimental scientists, are beyond the resources of individual efforts. Combining an investment in large, common infrastructural components with the existing network of resources and practitioners will create an "HTE materials superhighway," increasing scientific impact and accelerating the development of new commercial technologies. This superhighway can be mobilized to address the urgent, high impact, and materials-constrained challenges facing the nation. Individual topics will reflect the diversity of each field; for example, novel transparent conductors encompass light-emitting diodes, solar cells, and smart windows.

Shared centers with core capabilities and expertise must be established to achieve this materials superhighway. These high-throughput experimentation centers (HTECs) should have on-site library synthesis, processing, and structural characterization capabilities, as well as staff with expertise in experimental design, fabrication, analysis, data management, and materials informatics. Furthermore, since structural and compositional analyses are the foundations of materials exploration, advanced characterization facilities such as synchrotron beam lines provide an ideal launching point for HTECs.

Such facilities will make HTE methodologies available to the larger research community and increase the quantity and statistical significance of data by enabling data sharing in digital formats. HTECs will also provide venues for experimentalists and theorists to collaborate and for materials informatics platforms and analysis software to be deployed to diverse datasets. It is critical that the resources of the HTECs are readily accessible to industry.

It is important to point out that there are currently no national facilities in which a comprehensive, sustained (one- to five-year effort) HTE approach to novel materials discovery and commercialization can be achieved. Although most elements of the required infrastructure exist, distributed over several government, academic and industrial enterprises, there are few mechanisms for interaction. Similarly, no single center (either brick and mortar, or virtual) can accommodate the breadth of HTE methodologies for all technology focus areas.

HTECs offer efficient use of resources through the establishment of shared techniques and facilities. Some elements, notably data tools and management, are common to all efforts. Many library synthesis tools can produce libraries of several materials classes, spanning a number of technologies. Highly specialized and expensive HTE characterization tools, such as x-ray diffraction beam lines, are valuable for research in virtually all materials classes. Thus, a system for sharing capabilities and minimizing duplication is required.

Geographically delocalized multi-institution centers that operate on five-year horizons with annual planning are needed. The ideal structure is likely the creation of national centers dedicated to using HTE approaches for specific materials-constrained technological grand challenges. These centers have to be adequately staffed with experts in synthesis, metrology and data management and contain the tools required to address the relevant technological challenges. The centers must also have access to a distributed network of research tools which exist in different laboratories (academia, national laboratories, and industry) around the world. Since it is not practical (or even possible) to have every synthesis, processing and metrology tool under one roof, a series of pre-arranged relationships to access a variety of tools on an as-needed basis must be created. Because the purpose of the centers is to provide materials solutions that can be quickly inserted to existing technologies, the equipment should be manufacturing grade. For example, centers should have facilities for thermal/reactive processing and measurement of HTE libraries at different atmospheres and temperatures. The goal of the HTECs may be described as “lab to fab.”

Due to the very nature of the HTE methodology, we envision the center facilities to serve as vehicles where a team of scientists and engineers can utilize the available tools to quickly get to the root of a particular problem and rapidly develop the necessary solutions for implementation (**Figure 3**).

HTE Case Studies and Success Stories

Dow Chemical Company had significant progress in the computational simulation, modeling, and predictions of materials properties. However, the synthesis of target materials often involves competition between a number of phases, indicating the need to determine a unique reaction pathway to produce and commercialize the desired phase. Dow found that HTE methodologies were uniquely suited to rapidly generate experimental data, providing effective tools to identify materials synthesis routes.

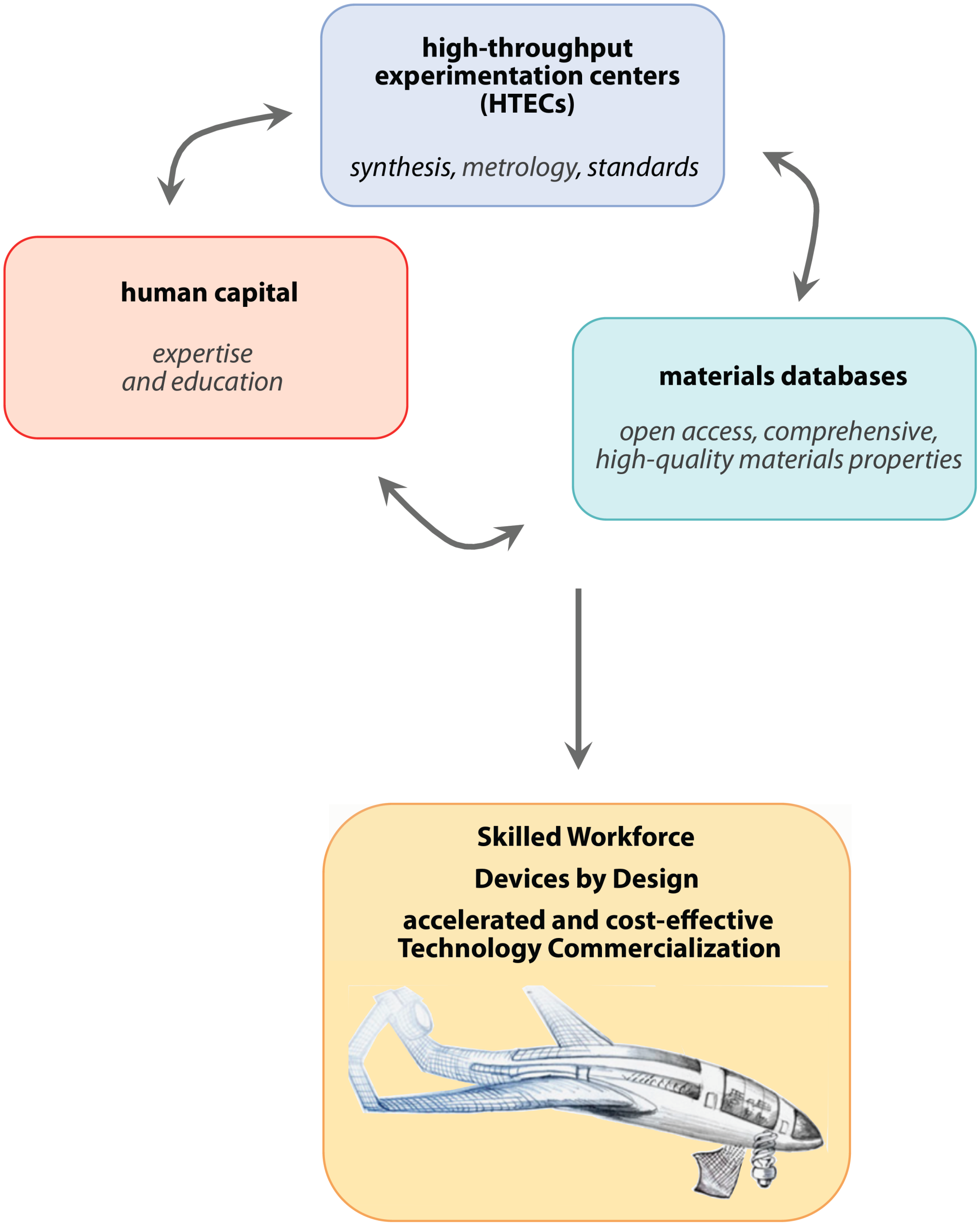

Figure 3. Developing solutions for significant challenges through a national effort in high-throughput experimentation. In combination with the growing advances in theory and computational materials modeling, HTE-generated experimental data will fulfill the promise of the MGI through fueling materials innovation and commercialization.

One such story highlighting Dow's successful use of HTE for new materials is the development of InFuse™ olefin block copolymers (OBCS). This two-year project produced new theoretically predicted materials, optimized production, and rapidly commercialized a new class of polymers now made at scales exceeding 10^8 pounds per year. OBCs had been known to possess desirable properties, but a cost-effective production technology had not been discovered. Theory showed that the geometry around a catalyst active site dramatically affects the incorporation of hard or soft segments in the polymer. The solution was therefore to develop a technology to shuttle the growing polymer chains between different catalysts with desired geometries. The selection process began with representative examples from a broad variety of catalyst structure types with high polymerization rates. The catalysts were then used in combination with numerous potential chain shuttling agents for ethylene polymerization reactions. Using automated polymer synthesis and characterization methods, more than 1600 individual polymerizations were evaluated over a three-week period, a feat that would have taken several months using conventional techniques. Soon afterward, the materials were swiftly commercialized and now represent a rapidly growing field of polymers with uses in artificial turf, plastic toys, and commercial carpet tiles. Such examples are illustrative of the role HTE methodologies will continue to play in the experimental realization and commercialization of materials developed in silico[12].

As a second case study, Intermolecular is an HTE services corporation that has worked extensively with the semiconductor industry, one of the largest consumers of advanced materials in the world. While the majority of Moore's Law achievements have been driven by advanced lithography, the importance of advanced materials has skyrocketed over the past 10 years, now accounting for more than 95% of the cost and effort that goes into developing products dictated by the semiconductor roadmap. As the integration of new materials and device architectures make up an ever-increasing proportion of overall device performance, the number of materials and materials combinations that semiconductor manufacturers must work with is growing rapidly.

The typical development cycle to move to the next node of Moore's Law is around 18 months. This is now largely driven by the final and most important stage of the typical MGI-targeted 10-20 year cycle: the development and integration of advanced materials into a high-value end product. The powerful economic impetus to shorten this time and reduce the cost for each new generation of semiconductor products is leading the semiconductor industry to be an early adopter of HTE, with significant success.

For example, Elpida Memory (now Micron Technology, Inc.) is a global leader in DRAM chip manufacturing for the consumer electronics industry. Their typical development cycle was around 18 months for each new product, but this pace became difficult to maintain as the challenges of incorporating advanced materials became more significant. In 2009, Elpida and Intermolecular began a collaboration to solve this problem, ultimately accelerating the time for each Moore's Law generation from 18 months to less than 6 months. To accomplish this, Intermolecular used specialized HTE fabrication and characterization tools to perform a series of high-throughput screens. Each screening step produced comprehensive data on materials, process, and device integration, enabling an increasing focus on higher probability material targets. The increased R&D efficiency, reduced time to production and reduced technical risk associated with the HTE approach may result in hundreds of millions of dollars in additional revenue per product generation. The DRAM products developed through this collaboration are now included in the iPhone 5.

Need for a Strong Educational Component

The highly diverse nature of university-based materials exploration efforts makes the participation of academic institutions critical to the success of the HTECs. Universities provide the training grounds for a diverse and flexible MGI-ready workforce capable of coalescing workflows involving theory, experimentation, and data science. A consortium of academic HTE experts will collaborate to develop a number of open access educational resources: web-based video tutorials on experimental best practices, experimental and data-driven materials courses, MGI lectures, and MGI-based virtual "labs" based on emerging materials problems.

There is a substantial educational opportunity embedded in HTE methodologies: the ability to train a new generation of materials scientists in the use of HTE techniques, libraries and data science. Creating opportunities for students and professors to contribute in industry and government labs will further increase their understanding of and effective research on devices, products and manufacturability; promote commercialization of university research; and provide academic expertise in industrial settings. Participation by professional societies (student chapters, workshops, and networking activities) and Federal support will enhance these activities. The expedited "lab to fab" concept will only be viable by including structured educational opportunities at all levels between materials discovery and commercialization.

Path Forward

The following strategic action items will position high-throughput experimentation (HTE) to contribute significantly to accelerated materials discovery and commercialization:

1) Identify critical technologies that are currently materials-constrained. Potential grand challenges include cost-effective batteries, ultrahigh-temperature metals for turbines in next-generation natural gas power plants, low-cost and high-efficiency solar cells, low-power multiplexed sensors for flexible electronics, CO_2 hydrogenation and selective methane oxidation catalysts, chemical and biological sensors, and reliably available substitute materials for critical elements.

2) Establish key specific targets for desired materials properties and performance, and fund centers with dedicated teams and infrastructure for tackling such specific materials topics, which could have immediate impacts (within three years).

3) Enable user access to high-throughput experimentation centers (HTECs) through a proposal process. A flexible array of manufacturing grade HTE tools should be available to the larger community, especially small companies and startups with seed investments and the need for rapid progress.

4) Ensure that data science and management are integral to the HTECs. All data sets and databases should be interoperable and use common formats, analysis protocols and informatics platforms.

5) Establish HTE standards for the centers. Standardized R&D platforms (both physical and virtual) for testing libraries and metrology tools will be required. Standard library samples must be developed.

6) Fund new HTE metrology development at the HTECs to tailor experiments for target industrial applications. HTE versions of standard characterization tools, such as x-ray fluorescence, micron-scale x-ray beams, atom probe, XPS, in situ synthesis monitoring, TEM and materials property characterization techniques must be developed.

To start quickly and effectively, the national HTE effort can capitalize on universities, national laboratories and industry facilities that already have extensive existing HTE tools and expertise.

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