Progress in High Temperature Materials and Systems in the U.S. DOE Nuclear Hydrogen Initiative

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INTRODUCTION

Nuclear powered hydrogen production can be performed today using liquid water electrolysis. With this technique, the nuclear power plant supplies electricity to the electrolysis cell, which splits water electrochemically into hydrogen and oxygen. The cost of hydrogen produced by liquid water electrolysis is not yet economically competitive with hydrogen produced from the reforming of fossil fuels, and other methods are sought for making hydrogen that can take advantage of the nuclear power source (no greenhouse gas emissions, secure energy supply, reliability) at a lower cost.

Hydrogen production techniques currently being considered for nuclear-powered hydrogen production can be divided into two classes: thermochemical methods, and electrochemical methods. Though both classes use water as a hydrogen source, they differ on how the nuclear reactor is used to drive the water-splitting reaction. Thermochemical methods use the thermal energy from the nuclear reactor to drive multi-step cyclic chemical reactions to perform the water-splitting task. Alternatively, electrochemical methods rely primarily on the use of electricity generated by the nuclear reactor. There is a subset these two classes, called thermochemical hybrid methods, which use thermal energy for some reaction steps, and electricity for others. These classes are represented in the DOE Nuclear Hydrogen Initiative by the Sulfur-Iodine method (Ref. 1), High Temperature Electrolysis (Ref. 2), and the Hybrid Sulfur method (also known as the Westinghouse Sulfur Cycle, Ref. 3), respectively.

Common to all of these methods is the need for high operating temperatures, 700-1000 °C, in order to achieve higher process efficiencies. Since water splitting is an endothermic reaction, these hydrogen production processes are unable to supply their own thermal energy, and the nuclear power plant must supply the thermal energy to drive the water-splitting processes. The thermal energy delivered must also be of high enough quality because water is very thermodynamically stable and will not readily decompose into its constituents until the temperature exceeds 2000 °C. The current generation of nuclear power plants operates at temperatures no higher than 300-350 °C, and higher temperature nuclear reactors, as being developed under the U.S. DOE Generation IV Nuclear Energy Systems Initiative, will be needed to drive these hydrogen production methods.

Due to safety concerns, nuclear plants and hydrogen production plants will not likely be integrated into a single unit, and will instead be separated by physical barriers and distance that will protect the nuclear plant in the event of an accident at the hydrogen production plant. This physical separation makes transmission of thermal energy from the nuclear plant to the hydrogen production plant a significant challenge. High temperature heat exchangers and a high temperature heat transfer loop must be developed that is capable of transmitting high-temperature thermal transfer fluids over significant distances (tens to hundreds of meters) with minimal heat losses in order to provided energy to the hydrogen production processes (see Figure 1). Collectively, the heat exchangers and the heat transfer loop are called the System Interface. Development of the System Interface is one of the focus areas of the System

Interface and Support Systems area under the U.S. DOE Nuclear Hydrogen Initiative, and is the area of interest in this paper.

Significant technical barriers to the construction of the System Interface exist in the areas of materials, heat exchanger design and construction, system design and control, and in loop operation (Ref. 4). There are considerable materials demands on the thermal energy transfer loop because it must simultaneously contact the high-temperature coolant used in the nuclear reactor, and contact the relatively cooler but more corrosive fluids used in the hydrogen production processes (e.g., sulfuric acid, steam/hydrogen). The heat transfer loop must employ a high temperature/pressure gaseous heat transfer fluid (e.g., helium, helium/nitrogen) or a lower pressure liquid heat transfer fluid (molten salt) to move the thermal energy, which may cause mechanical failures due to creep or corrosion. Advanced heat exchangers using appropriate materials must be designed and constructed that can reconcile the needs for high-temperature strength, creep and corrosion resistance, while keeping within cost goals. Also, careful study of loop control and operation must be performed to ensure that it will operate in a very stable manner without readily communicating or propagating plant upsets through the heat transfer loop.

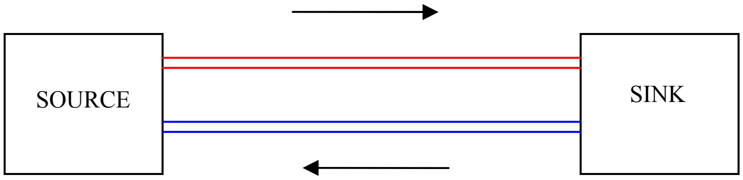

Figure 1: Nuclear plant (source) connected to H_2 plant (sink). Top line shows flow of hot fluid towards H_2 plant and bottom line shows flow of cold fluid back to the nuclear plant.

This paper provides an overview of the technical progress made so far under the U.S. DOE Nuclear Hydrogen Initiative in the development of the thermal energy heat transfer loop for application to nuclear powered hydrogen production. An integrated research program has been initiated in this area that involves the cooperation of national laboratories, universities and private entities. Work has been accomplished in high temperature materials property measurements, corrosion behavior, heat exchanger designs, nuclear plant/hydrogen plant separation distance needs, system interface configurations, and identification of heat transfer fluid candidates. Near-term research plans (1-3 year time frame) and will also be identified.

MATERIALS

The design of a high-temperature heat transfer network will require the use of high-temperature metallic alloys, and most likely the judicious use of ceramic or ceramic composite materials due to the high temperatures required (900+ °C), high pressure differentials (0-7 MPa), and the use of corrosive or reactive chemicals in the hydrogen production processes (sulfuric acid, hydrogen iodide, steam/hydrogen mixtures, etc.). Though the properties of most of the materials of interest are generally known, it is important to determine where gaps in the materials properties databases exist and fill in those gaps, and to determine how the materials will behave in regard to crack formation, creep, and corrosion under the specific circumstances required for construction and operation of the System Interface. Work on measuring materials properties has been ongoing at the University of Nevada Las Vegas (UNLV), University of California Berkeley (UCB), the Massachusetts Institute of Technology (MIT), Ceramatec Inc. in Salt Lake City, UT, and at General Atomics (GA) in San Diego, CA.

METALS

For metals, the properties of interest are the mechanical properties at temperature (yield strength, modulus, toughness, creep), degradation behavior (general corrosion, localized corrosion, stress corrosion cracking, crack growth rates, hydrogen embrittlement effects), and changes that occur over time in the microstructure. The metals currently under study for general structural applications are Hastelloy C-22, Hastelloy C-276, Waspaloy, Inconel 800H, and Inconel 617.

At UNLV, the tensile properties of Hastelloys C-22 and C-276, Waspaloy, and Inconel 800H have been measured up to 600 °C and compared to literature data, while at the Idaho National Laboratory, the tensile and crack behavior properties of Inconel 617 have been measured up to 1000 °C. Of these metals, Inconel 617 shows the highest strength and creep resistance above 800 °C in comparison to the other metals in the literature, and is already being considered for use in the heat exchanger joining the nuclear reactor to the System Interface. Work is underway at UNLV to expand the temperature range of study up to 1000 °C for the remaining alloys.

Stress corrosion cracking behavior of Hastelloy C-22, Hastelloy C-276, and Waspaloy in aqueous solutions of sulfuric acid and sodium iodide at 90 °C and at constant load and slow-strain-rate have been performed, and equivalent tests with Inconel 800H have been initiated. Specimens exposed to the test solutions have been examined by scanning electron microscopy. Aqueous sulfuric acid and sodium iodide solutions are representative of some of the chemical conditions that may be encountered in the Sulfur-Iodine process. The tests are still ongoing, and there are no conclusions yet regarding which material has the best performance.

For specific application to the hydrogen iodide decomposition step in the Sulfur-Iodine process, refractory metals and refractory alloys have been studied at GA. Fourteen metal specimens were immersed in HI_x ($HI + H_2O + I_2$) solutions at various concentrations up to 310 °C for time periods of 100 hours or more to watch for gross changes in the materials (changes in color, sample dimensions, and mass) and to screen for materials failures. Metals and metal alloys showing very good corrosion resistance to the test solutions include Ta, Nb, and their alloys with tungsten and hafnium. Zr, Mo, Hastelloy C-276, and Haynes 188 performed poorly and exhibited pitting or material dissolution. Three refractory alloys were selected for further research for this application because of their availability and relatively lower cost – Ta-2.5W, Ta-10W, and Nb-10Hf. Some photo results are shown in Figure 2.

Figure 2: Zr-705 after 120 hours, Ta-2.5W after 1040 hours in HI_x. Zr-705 shows visible corrosion, while Ta-2.5W shows no corrosion.

Unique alloys blending Inconel 800H plus Pt and Inconel 617 plus Pt have been developed at MIT for possible application to a metallic heat exchanger for the sulfuric acid decomposition step of the Sulfur-Iodine process. The alloys, containing 1-2 wt% platinum in the bulk and concentrated mainly on the exposed surfaces (surface impregnation), will be used to construct prototype heat exchangers that may be able to catalyze the chemical step $SO_3 \rightarrow SO_2 + \frac{1}{2} O_2$ without having to use solid-supported platinum to catalyze the decomposition reaction. So far, small samples have been generated and examined by optical metallography, and a chemical kinetics test apparatus has been constructed to measure the catalytic activity of the samples. If the material is sufficiently catalytically active and the platinum is stable on the exposed surfaces, a combined heat exchanger/chemical reaction unit may be developed without having to add additional catalyst. Issues such as effective catalyst surface area and a determination of catalyst stability and deactivation effects will be studied in a catalyst test apparatus being prepared at MIT and in test equipment located at the INL (Ref. 5).

CERAMICS

Ceramatec Inc. has selected sintered SiC, CVD-SiC, $MoSi_2$, Ti_3SiC_2, Si_3N_4, Al_2O_3, SiAlON, and cordierite ($2\ MgO - 2\ Al_2O_3 – 5\ SiO_2$) as candidate ceramic materials for application to a high temperature ceramic heat exchanger for the sulfuric acid decomposition step of the Sulfur-Iodine process. Available literature data indicate that these materials may have the most favorable properties for this application in regard to mechanical behavior and corrosion resistance. A test apparatus has been constructed that will be capable of exposing stressed and unstressed specimens of these materials to mixtures of steam, sulfuric acid, and oxygen at temperatures up to 950 °C, and will be used to collect data on these materials in the coming year.

At UCB, carbon fiber reinforced SiSiC (FR-SiSiC), splint based SiSiC (SB-SiSiC), and pitch based carbon fiber reinforced SiSiC (BioKer) composite materials are being investigated for application to high temperature compact heat exchangers that may be used for the nuclear reactor/system interface heat exchanger or the sulfuric acid decomposition reactor. Coupons of these materials have been prepared and tested to measure their mechanical properties. The measured mechanical properties will be used to improve numerical models of compact heat exchanger configurations. Coupons of these materials have been manufactured for corrosion testing and to study manufacturing methods for larger structures.

An independent project is underway at Johns Hopkins University to investigate cellulose-derived SiC for application to compact heat exchangers. Dense SiC monoliths have been formed through the Liquid Silicon Infiltration process having apparent densities of up to 96.0% of theoretical density. These materials will be examined with BET, XRD, and SEM to study the microstructure of the material and to seek ways to improve the quality of the material and the manufacturing methods in the coming months.

General Atomics performed corrosion testing on ceramic materials exposed to HI_x solutions, as was done with metallic samples (Ref. 6). Their testing showed that sintered SiC from Ceramatec, CVD-SiC, Bioker SiSiC, and SB-SiSiC performed well with no evidence of corrosion in the test solutions, while FR-SiSiC and alumina showed evidence of corrosion after 100 hours of exposure. Another ceramic material, mullite ($3\ Al_2O_3\ 2\ SiO_2$) also showed very good corrosion resistance to HI_x solutions and may provide a lower cost alternative for some heat exchanger components in comparison to the SiSiC composite materials for this application. Extruded graphite was also tested and found to be corrosion resistant but porous, and the test samples were observed to absorb the test solution.

HEAT EXCHANGER DESIGN AND DEVELOPMENT

UNLV, MIT, and Ceramatec Inc. are exploring compact heat exchanger designs for application to sulfuric acid decomposition in the S-I process. Computational fluid dynamics and mechanical/thermal stress calculations of compact off-set strip fin ceramic plate heat exchanger designs are being performed to determine the optimal channel dimensions and other physical characteristics. Ceramic and metallic compact heat exchangers are under consideration, including concepts offererd by Ceramatec Inc. for a ceramic sulfuric acid decomposition heat exchanger, and a self-catalyzed metallic heat exchanger concept offered by MIT. Parametric studies have been performed to evaluate baseline heat exchanger operation by varying strip fin thickness, gap length, flow channel height, fin length, and pitch. The software packages FLUENT, ANSYS, and MATLAB have been used to study the thermal-hydraulics of the flow channels and to analyze thermal and mechanical stresses of the designs under examination. Sizing studies have been performed to optimize the size of heat exchanger modules and to determine how many modules would be needed for various power throughputs. Advances have also been made in how these numbers are computed, and parallel computing algorithms and systems have been established to more rapidly perform the calculations.

Highly compact heat exchanger designs that can be constructed from melt-infiltrated carbon-carbon composite heat exchangers and SiSiC composites are being studied at UCB.

At this stage of the research, UCB has manufactured molds with embossed features resembling the detailed flow channels that would exist in the actual heat exchangers. These molds will be used for lamination and polymer infiltration and pyrolysis (PIP) processing of materials to construct sample heat exchanger plates and coupons for study. The purpose of this work is to determine the physical properties of the individual plates and to more fully develop the manufacturing methods, so that prototype heat exchangers may soon be constructed.

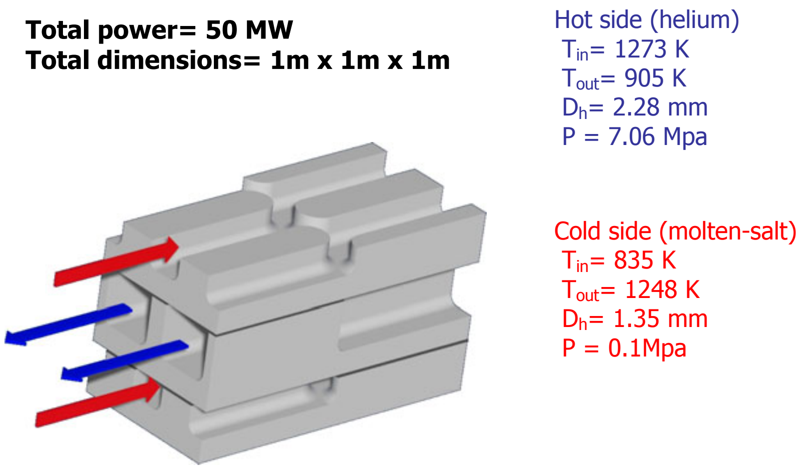

Figure 3: Example of compact strip-fin ceramic heat exchanger using helium on the hot side and molten salt on the cold side.

UNLV is constructing a heat exchanger testing facility that will allow for the testing of prototype heat exchangers under scalable fluid conditions. The facility will be used to benchmark thermal-hydraulic data and thermal properties of fluids in order to improve heat exchanger models. Helium or inert gas mixtures can be used to study gaseous heat transfer fluids, and silicone oil can be used to represent the fluid properties of molten salts. Single-fluid and double-fluid test channels composed of aluminum alloy 6061 have been constructed that will be used to mimic individual flow channels and plates in metallic and ceramic compact heat exchangers. Fluid conditions have been chosen to resemble the conditions that would be experienced in the nuclear reactor/system interface heat exchanger, assuming helium is the hot fluid and molten salt is the cold fluid, based on a comparison of Nusselt, Prandtl, and Reynolds numbers (see Figure 3). The heat exchanger testing apparatus design and operation draws on work performed at UCB (Ref. 7) in which a similar apparatus was constructed and used to perform scaled thermal hydraulics experiments for liquid salt and helium systems.

NUCLEAR PLANT/HYDROGEN PLANT SEPARATION

From an energy efficiency and thermal transmission viewpoint, the best arrangement is to co-locate the nuclear plant and the hydrogen plant, so that the two plants are integrated into a single facility. From a safety and regulatory standpoint, such a co-location is not acceptable, and the hydrogen plant must be located sufficiently far away from the nuclear plant to prevent damage to the nuclear reactor in the event of an accident at the hydrogen plant. It is well known and anticipated that the hydrogen plant will have a higher probability of accidents and equipment failures due to the presence of corrosive chemicals, purified oxygen, hydrogen, and high temperatures. Also, the overall combined facility must be designed to handle those events without causing anything worse than localized damage to the facilities.

An initial effort has been made to define the minimum plant separation distance by examining the frequency of hydrogen accident events at nuclear plants, the existing sets of safety rules from the U.S. Nuclear Regulatory Commission (NRC), Occupational Safety and Health Administration (OSHA), National Fire Protection Association (NFPA), and the American Society of Mechanical Engineers (ASME), and the projected failure modes of equipment in the proposed hydrogen production process designs, and then using the tools of probabilistic analysis to generate a range of acceptable distances (Ref. 8). Failure fault trees were developed using random hydrogen explosions, human error, and seismic events as initiating events, and assigning probabilities to the consequences of those events. Chemical dispersion modeling was also performed. The minimum distance required between the plants was determined by increasing the spacing between the plants as a function of the failure probabilities, presence or absence of physical barriers, above/below grade position of the plants, and so forth, until the probability of nuclear core damage fell below 1×10^{-6} events/year, the maximum probability of core damage above which events, as regulated by the NRC, become greater than “extremely unlikely.” The SAPHIRE model was used to analyze the fault trees, and the ALOHA code was used to examine chemical dispersion (excluding hydrogen dispersion).

The conclusions from this study were that a minimum separation distance of 60 to 120 m is required to protect the nuclear plant core from damage resulting from accidents at the hydrogen production plant. The inventory of hydrogen stored at the hydrogen plant should be minimized as much as possible to reduce the magnitude of any hydrogen explosions that do occur. Siting of the nuclear reactor control room was listed as a significant safety challenge because dispersion of hazardous chemicals (sulfuric acid, sulfur dioxide, etc.) affects a much larger area than would hydrogen explosions and can affect reactor control personnel without causing nuclear reactor core damage.

It is recognized that this report is only the beginning of a series of revisions and studies on overall system studies, and it will be necessary to revise and update this information as the nuclear plant designs and hydrogen production technologies become more defined.

SYSTEM CONFIGURATION AND OPERATION

Thermal-hydraulic studies of system interface configurations have been performed to determine the effects of choosing different heat transfer fluids and of different nuclear reactor power conversion cycles on the temperature drop and energy losses across the heat transfer loop (Ref. 9). Seven possible configurations for a system that transfers heat between the nuclear reactor and the hydrogen and/or electrical generation plants were identified (see Figures 4 and 5 for examples). Both helium and two molten salts (FLiNaK and $NaBF_4$-NaF) were considered as the working fluid in the heat transport loop. Methods were developed to perform thermal-hydraulic and cycle efficiency evaluations of the various components in the heat transport loop for the various configurations. The relative sizes of components provided a relative indication of the capital cost associated with the configurations. Estimates of the overall efficiency of the configurations were determined. The efficiency calculations were used to determine which heat transfer fluids and power conversion configurations are the most promising from a thermal-hydraulic and efficiency points of view.

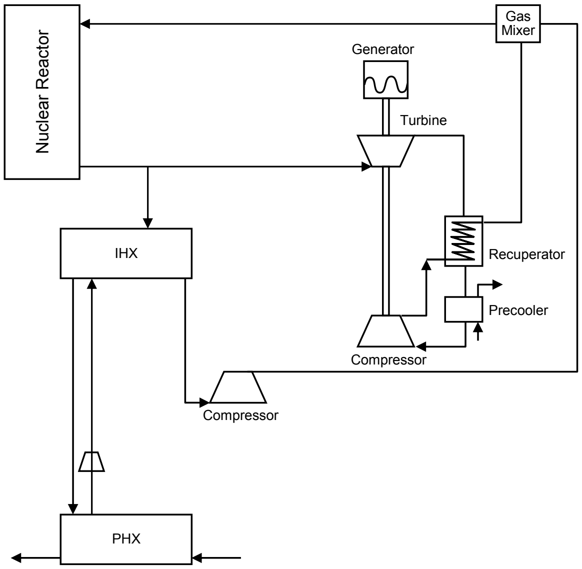

Figure 4: Direct electrical cycle and a parallel intermediate heat exchanger (IHX). System interface lies between the IHX and the PHX.

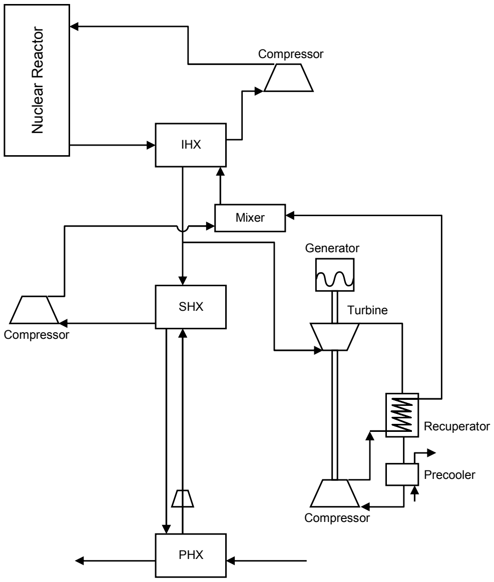

Figure 5: Indirect electrical cycle and a parallel secondary heat exchanger (SHX). System interfaced lies between the SHX and the PHX.

In addition to efficiency and heat loss calculations, mechanical stresses on piping were also examined, and recommendations were made regarding system operating pressures and temperature limits. It was assumed that the heat transport loop was constructed of Alloy 617, that the heat transport pipes were arranged in a side-by-side parallel arrangement (as opposed to a concentric arrangement), and that the receiving heat exchanger in the loop was a shell-and-tube heat exchanger at the hydrogen production plant.

There were a number of conclusions from this work. The stress and creep rate data suggested that the maximum operating pressure of the heat transfer loop would be 2 MPa, as determined by the mechanical limits of a shell-and-tube heat exchanger constructed of Alloy 617 and operating at a temperature of 900 °C. Higher pressures can be used, but the tube thickness becomes excessive in this heat exchanger due to the need to prevent creep-based heat exchanger rupture in less than 10^5 hours. Compact heat exchangers would allow for higher operating pressures at this temperature. Assuming helium is the heat transfer fluid, pipe diameter and compressor power can be reduced by 30% and 20%, respectively, if the system pressure is increased from 2 MPa to 7 MPa. Indirect energy conversion cycles are much safer in terms of preventing cross contamination and achieving isolation between plants, but theyincrease thermal losses and loop volumes by up to 60%. Using a liquid salt heat transfer fluid allows for lower system pressures and reduces pumping power by 30-60%. Also, piping diameters are reduced by about 80%. Salts have disadvantages, however, in that they have high freezing points, and the time to freezing with a loss of loop power was on the order of minutes to hours.

Recommendations were drawn from these results. The use of a low-pressure fluid allows for relatively simple mechanical design for the loops because metallic alloys could be used. The heat exchanger in the intermediate heat transport loop should be arranged in parallel rather than in a serial arrangement with the electrical conversion unit. The best configuration options in terms of efficiency and temperature losses were the following: direct electrical cycle with parallel intermediate heat exchanger, direct electrical cycle with parallel intermediate and secondary heat exchangers (for added isolation between the nuclear reactor primary cooling loop and the hydrogen plant), and indirect electrical cycle with parallel secondary heat exchanger. Liquid salts should be explored further for use in the heat transfer loop because of their increasing benefits at larger plant scales in comparison to compressed helium.

An additional study was performed that examined the implications of a concentric arrangement of the hot and cold pipes, with the hot supply pipe nested inside the cold return pipe (Ref. 10). The study concluded that a concentric arrangement is superior to a side-by-side arrangement in terms of heat losses and in lowering the pressure differential (and mechanical stresses) on the hot supply pipe, but may be more difficult to manufacture and repair than a parallel pipe arrangement if a leak or failure of the inner hot supply pipe occurs. This study also determined that mechanical stress limits may be exceeded in the standard Heatric heat exchanger arrangement composed of Alloy 617 at 900 °C at for pressure differentials exceeding 3 MPa. Alternative heat exchanger configurations are proposed for further study that would reduce the probability of localized failures due to stress concentrations.

FUTURE WORK

The studies performed so far in the research effort are part of an overall strategy to develop the technology that will enable the construction of a pilot-scale heat transfer loop as part of a hydrogen production process demonstration employing one of three hydrogen production technologies -- Sulfur-Iodine, HTE, or Hybrid Sulfur. Figure 6 shows the path being taken to reach this goal.

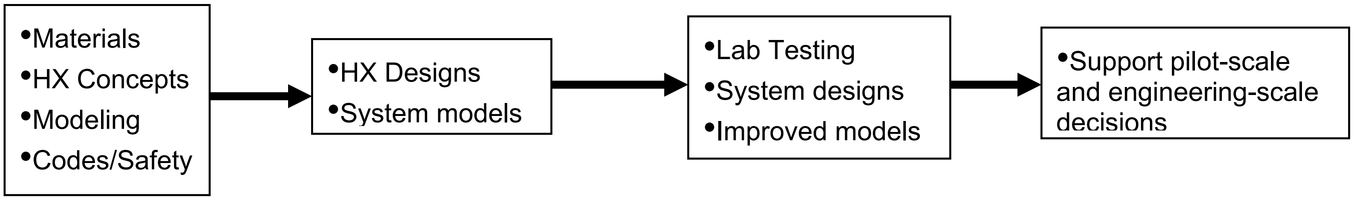

Figure 6: System interface development pathway.

Parallel projects are being pursued now in the areas of materials, heat exchanger design and development, system modeling, and in the area of codes and safety. Information from these areas will be used to down-select specific heat exchanger designs and more accurately develop system models. Then, prototype heat exchangers can be constructed out of the appropriate materials and tested in the laboratory, and the system models can be improved still further with laboratory data. All of this information will then be used to support design and construction decisions for pilot-scale equipment (10 kW to 1 MW power range) and, eventually, engineering scale equipment (50 MW).

Within the current DOE NHI timelines, heat exchanger prototype testing is slated to begin in 2007, and models capable of simulating the steady-state and dynamic behavior of the entire system interface as it interacts with the nuclear plant and the hydrogen production plant will be completed in 2008. Materials testing and revision of heat exchanger designs as laboratory data and improved models become available will be an ongoing activity. In 2006, the plant spacing requirements will be re-assessed in 2006 to include chemical plant accident data. Also in 2006, a full assessment of applicable codes and standards to a hydrogen production plant and system interface will be performed.

Cooperation between the national laboratories, participating universities and private entities will continue, and collaborative research will be emphasized in order to maximize the amount of research and development that can be performed within the funding limitations of the program.

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