II.D.5 Bulk Amorphous Hydrogen Purification/Separation Membranes

Thad Adams
Savannah River National Laboratory (SRNL)
773-41A/151
Aiken, SC 29808
Phone: (803) 725-5510; Fax: (803) 725-4553
E-mail: thad.adams@srnl.doe.gov

DOE Technology Development Manager:
Arlene Anderson
Phone: (202) 586-3818; Fax: (202) 586-9811
E-mail: Arlene.Anderson@ee.doe.gov

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Objectives

- Demonstrate the feasibility of using metallic glass materials in bulk form for novel advanced hydrogen purification membranes.
- Develop optimized bulk amorphous alloy compositions for hydrogen separation membranes.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Production section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

(A) Reformer Capital Cost
(C) Operation and Maintenance (O&M)
(F) Control and Safety
(N) Hydrogen Selectivity
(T) Capital Cost and Efficiency of Biomass Gasification/ Pyrolysis Technology

Technical Targets

DOE 2010 Separation Membrane Targets:

- Flux rate: 250 scfh/ft^2
- Cost: <$1,000/ft^2 (module cost including membrane material)
- Durability: 26,280 hours

Approach

Separation and purification membranes must have high hydrogen solubility, high diffusivity and catalytic activity on the surface of the membrane. Hydrogen separation as described in this work can be accomplished by the use of bulk amorphous materials (namely, bulk metallic glasses) permeable to hydrogen. The focal point of the SRNL effort will be on the development and optimization of a bulk amorphous material for the dense metallic-based membrane substrate.

The current generation of gas separation membranes is based on palladium/palladium alloys used either independently or in conjunction with porous ceramic supports. Pd/Pd alloys have been known to possess the ability to dissolve considerable amounts of hydrogen and to demonstrate increasing permeability with increasing pressure differential and temperature. However, the major drawbacks to their industrial use are the high cost for Pd, the relatively low flux, and that during cycling above and below a critical temperature, an irreversible change takes place in the palladium lattice structure which can result in significant damage to the membrane. SRNL has previously worked with thin section (melt–spun ribbons) of metallic glass materials for membrane applications, however, with the relatively new ability to cast fully amorphous metallic glasses in bulk sections. As a result, a new opportunity has opened for bulk metallic glasses (BMG) as hydrogen membranes. The ability to readily cast BMG alloys will allow for easier fabrication of membranes – machine thin membranes from larger BMG casting – and will also ease mass production challenges in comparison to thin section (melt spun) metallic glass ribbons. BMG alloys are traditionally processed from multi-component systems comprised of metallic species of varying atomic size. It is this vast difference in atomic sizes that results in slow diffusion/redistribution kinetics and allows for deep undercoolings to the point of freezing in the "liquid" structure to produce amorphous metallic alloys at relatively slow cooling rates (10-100 K/s). These BMG alloys have been shown to possess high permeation rates. For example, the permeation rate for a Zr-Al-Co-Ni-Cu BMG alloy (1.13 x 10^{-8} mol/m s Pa$^{1/2}$) is comparable to the permeation rate measured for pure Pd metal. Furthermore, these BMG alloys have also been shown to possess high elastic toughness and excellent resistance to hydrogen degradation. Both of these properties, high permeation and high elastic toughness, potentially makes these materials attractive for gas separation membranes.

Accomplishments

Funding to support this project did not begin until May of 2007. As a result, the initial focus has been to identify commercial available metallic glass product forms for testing and to acquire these materials. To date, materials from three different alloy systems have been acquired and are as follows: 1) Zr-Cu-Ni-Al-Y bulk metallic glass plate from Howmet Research, 2) Fe-based metallic glass film from MetGlass Inc, and 3) Co-based metallic glass film from Metglass Inc. As an initial screen of these alloy systems for hydrogen permeability, high throughput electrochemical permeation testing following ASTM standard G-148 has been initiated. This test technique provides a safe, simple method for screening multiple alloy systems prior to conducting gaseous hydrogen testing. The following accomplishments have been made:

- Acquired metallic glass materials from commercial vendors.
- Set-up Devanathan-Stachurski electrochemical hydrogen permeation test cell.
- Ordered Gamry potentiostat for electrochemical testing.
- Drafted test plan and finalized testing hazards analysis package.