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Hydrogen inactivation of liquid metal heat pipes

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ABSTRACT

When heat pipes are applied in atmospheres containing hydrogen, e.g. in presence of syngas, a small amount of hydrogen permeates through the wall of the containment into the heat pipe and accumulates as non-condensable at the end of the condenser. If the hydrogen is not removed during operation, this mechanism causes a rapid deactivation of the heat pipe starting from the condensation zone. This paper demonstrates the impacts of this deactivation on heat pipe operation and heat transfer rates and presents a formal description of the mechanism. Possible countermeasures to avoid this problem are presented and will be discussed, in particular the choice of safe operation conditions and the use of hydrogen windows based on metal membranes.

The hydrogen deactivation was investigated both experimentally and theoretically. We will focus on the kinetics of the inactivation process as well as the equilibrium state when applying hydrogen windows for hydrogen removal. The results will be discussed in comparison with modelling approaches. The deactivation time from beginning of hydrogen sweep to complete inactivation of the heat pipe is in the range of 1–3 h. In the experiments a nickel hydrogen window was applied in a sodium heat pipe. With this measure the inactive length could be limited to 25–31% of the heat pipe length in hydrogen atmosphere.

1. Introduction

Heat pipes are used for efficient and isothermal heating of processes in energy process engineering and in chemical industry applications requiring high heat fluxes. They consist of a metal housing containing a working fluid which can be evaporated in the so-called evaporation zone by applying heat. The vapour flows through the housing to the condensation zone where heat is removed causing the working fluid to condense. The liquid working fluid flows back to the evaporation zone driven by capillary or gravitational force. The system works isothermally as the heat transfer is only driven by evaporation. Ongoing research topics for the application of high temperature heat pipes at FAU-EVT are the carbonate looping process, biomass gasification using the Heatpipe Reformer and SOFC stacks with integrated heat pipes. For all of these applications sodium heat pipes with high-temperature steel containers are used.

In processes involving gaseous hydrogen the heat pipes deactivate due to hydrogen permeation to the inner side as shown in Fig. 1. A small amount of the hydrogen from the process permeates through the container wall into the heat pipe, accumulates at the cold end of the heat pipe and the heat pipe deactivates starting from the condensation zone. Due to the constant working fluid circulation the hydrogen partial pressure in the active part of the heat pipe remains zero and equals the vapour pressure of the working fluid in the inactive part. When applying a cooling duty, the inactive part has a much lower temperature than the rest of the heat pipe as no working fluid can condense in this region. Heat transfer only happens through conduction and diffusion with thermal resistances several degrees of magnitude higher than in the operating heat pipe. This phenomenon is called a “cold finger” [18].

We already described the heat pipe inactivation phenomenon in previous papers on the Heatpipe Reformer in [13] and thermal management of SOFC stacks using heat pipes in [5]. During the EU-project “BioHPR” (Ref. ENK5-CT-2000-00311) the inactivation was investigated by Groll et al., who examined different coatings like ZrO_2, Y_2O_3 and oxide layers to reduce hydrogen permeability of the housing [10]. Richardson et al. proposed a methane steam reforming reactor using heat pipes for heat transfer for isothermal operation and fast load changes [24,23]. However, in bench-scale experiments he encountered a complete inactivation of the heat pipe after 19 h of operation. The re-activation of the heat pipe took 28 days due to low the low temperature in the hydrogen-filled cold finger [24]. North and Anderson discussed heat pipe inactivation as a problem for the operation of heat pipes in bimodal space nuclear systems and proposed passive systems to remove the hydrogen from the heat pipe during operation [18,19]: For the construction of a heat pipe which can work in hydrogen containing atmospheres

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Nomenclature

Latin letters

A	surface area, m^2
c	concentration, –
D	diffusivity, mol m^{-1} s^{-1}
D	diameter, m
E_A	activation energy, kJ mol^{-1}
H	reduction factor, –
j	molar flux, mol m^{-2} s^{-1}
l	length, m
n	partial pressure exponent, –
$\dot{n}$	molar flow, mol s^{-1}
P	permeability, mol m^{-1} s^{-1} $Pa^{-0.5}$
P_0	permeability constant, mol m^{-1} s^{-1} $Pa^{-0.5}$
p	pressure, Pa
p_{LV}	vapour pressure, Pa
R	gas constant, kJ mol^{-1} K^{-1}
S	solubility, $Pa^{-0.5}$
t	time, s
T	temperature, K
x	*X*-coordinate, m
Δx	wall thickness, m

Greek letters

τ	time coordinate, s

Subscripts

az	active zone
g	gas
H	housing
HP	heat pipe
H_2	hydrogen
iz	inactive zone
l	liquid
MeOx	metal and oxide
Me	metal
W	hydrogen window

Abbreviations

HP	heat pipe
HPR	Heatpipe Reformer
MFC	mass flow controller
OLS	ordinary least squares estimation
SOFC	solid oxide fuel cell
SS	stainless steel

it is possible to either reduce the hydrogen flow into the heat pipe or enhance the flow leaving the heat pipe using a so-called "Hydrogen Window".

For the first approach the container should comprise of a material with a very low hydrogen permeability. Also, the operating temperature of the heat pipe plays an important role as the hydrogen permeation is strongly temperature dependent. Unfortunately, in most applications the operating temperature can not be chosen freely. It is also possible to use coatings which reduce the hydrogen solubility on the surface [8,18].

The only part where the partial pressure of the hydrogen inside the heat pipe is high enough for an efficient removal is the inactive part. The hydrogen flow out of the heat pipe is greatly diminished because of the lower temperature of the container due to the deactivation. The construction of the hydrogen window should therefore keep the temperature of the material as high as possible and use a material with a high hydrogen permeability, e.g. nickel alloys or palladium.

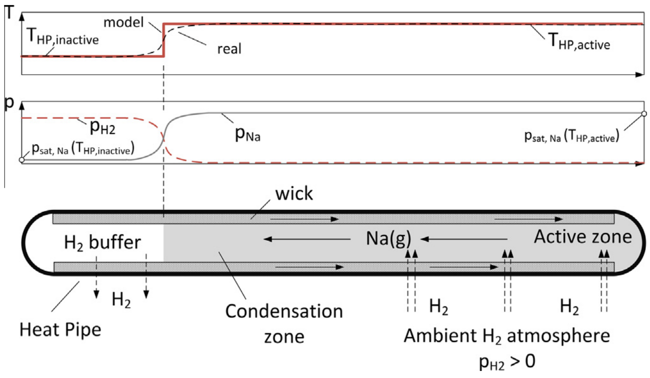

Fig. 1. Scheme of the inactivation of heat pipes due to hydrogen permeation.

2. Materials and methods

It is important to investigate the mechanisms of hydrogen transportation through dense materials as well as the working principles of heat pipes in order to design a hydrogen tolerant heat pipe. Heat pipes consist of the metal container for the heat transfer fluid, made of a steel pipe, a mesh structure on the inner side of the tube for the transport of the condensed fluid to the evaporation zone and the heat transfer fluid itself.

2.1. Fundamentals of heat pipes

Heat pipes are passive heat exchangers which rely on a evaporation–condensation mechanism: When heat is supplied to the one side the working fluid evaporates and flows to the cold end where it condenses. The condensate is transferred back to the evaporation zone by gravitational or capillary force. The operating pressure of the heat pipe equals therefore the vapour pressure of the working fluid p_{LV} at the respective operating temperature T_{HP}.

The heat transfer of a heat pipe can be limited by the properties of the working fluid, heat pipe geometry and capillary structure, these limitations are discussed thoroughly in [6]. For the following considerations it is assumed that the heat transfer is only limited by radiation and convection on the heat pipe outer surface and therefore the active length of the heat pipe.

2.2. Hydrogen permeation through metals

The permeation of diatomic gases like hydrogen through a dense body is divided into several steps: The adsorption of the hydrogen at the surface, the dissociation into protons and dissolution into the metal body, which is sometimes referred to as solution of the hydrogen. Then the diffusion through the metal in form of protons, which is in most cases the rate-limiting step of