

RECENT DEVELOPMENTS IN ADSORPTION MATERIALS AND HEAT EXCHANGERS FOR THERMALLY DRIVEN HEAT PUMPS

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Abstract:

Adsorption chillers and heat pumps are a promising technology for the supply of heating and cooling, especially in the small scale range. Therefore, adsorption materials with a high load capacity and heat exchangers with effective heat and mass transfer ratios are required. To compare the characteristics of adsorption-refrigerant working pairs from different manufacturers a common measurement method is required. This paper gives a literature survey about the state of the art in adsorption material developments and proposes a common measurement procedure for adsorbents. Moreover the latest activities in heat exchanger developments for adsorption heat exchangers and evaporators are represented.

Key Words: adsorption, heat exchangers, measurement procedure, evaporation, boiling

1 INTRODUCTION

In this paper an overview is given about ongoing research on thermally driven heat pump (TDHP) machines, especially adsorption TDHPs. In the first part the focus is on the adsorption material. Much research is performed in developing new materials and characterizing their physical properties. Yet, there is no common method or standard to measure and describe the adsorbent characteristics. Therefore, in the first section a proposal for a common measurement procedure is described.

The next question arises when adjusting the adsorbent to the heat exchanger. Different methods like coating and loose grain are available each with specific pros and cons. The target is to increase the heat and mass transfer for adsorption and desorption processes and at the same time increasing the adsorbent per heat exchanger capacity ratio.

When aiming for cheap and compact TDHPs a point is reached when the evaporator is the limiting component. In the last section improvements are shown to increase the heat transfer characteristics of the evaporator. The focus is to increase the heat transfer coefficients between the primary and secondary site of the evaporator and thus enhance the overall heat transfer mechanism between chilled water and refrigerant.

2 CHARACTERISATION OF ADSORPTION MATERIALS

For further improvement of adsorption type TDHPs it is of importance to identify useful working pairs, to measure their physical properties and to proof the stability. Many working groups in different research institutes are working in this field, focusing on different material classes and manufacturing processes.

Publications about the developments of new materials can be found e.g. at (Wang, Metcalf et al. 2012) and (Grekova, Gordeeva et al. 2012). Carbon materials in combination with methanol as refrigerant can be found e.g. at (Gordeeva, Freni et al. 2012) or at (Gordeeva, Freni et al. 2012; Henninger, Schicktanz et al. 2012; Schicktanz, Hügenell et al. 2012). The possibilities of metal-organic frameworks for water adsorption are described e.g. in (Henninger, Habib et al. 2009; Henninger, Jeremias et al. 2011; Henninger, Jeremias et al. 2012). Another material class are salt embedded frameworks such as described in (Sapienza, Glaznev et al. 2011). Further publications about adsorption materials are e.g. (Srivastava and Eames 1998; Aristov, Restuccia et al. 2002; Janchen, Ackermann et al. 2002; Critoph and Zhong 2005; Henninger, Schmidt et al. 2010).

To describe the performance of adsorption-refrigerant working pairs several physical properties need to be known, such as

- the ability of adsorption (equilibrium conditions)
- density
- thermal conductivity

It turns out that some of the working pairs although showing promising characteristics may lose stability with time. This is due to different mechanisms during the manufacturing process and the operation conditions. The materials therefore need to prove their stability against

- mechanical stress
- thermal stress under adsorption and desorption conditions (hydrothermal stability)

The hydrothermal stability of some materials are investigated e.g. in (Henninger, Munz et al. 2011) and (Freni, Frazzica et al. 2011) and (Freni, Frazzica et al. 2013).

In order to compare the measurement results and to find the working pair best fitting to a certain application it is of importance to have a common methodology for measurements. Such methodologies are described e.g. in (Henninger, Freni et al. 2011) with further improvements as described in (Henninger, Freni et al. 2012). Within the next section a procedure is proposed.

2.1 General overview

Three possibilities exist to measure the ability of adsorption materials each with their individual pros and cons. The most frequent measurement method used is thermogravimetry (TGA) and differential thermogravimetric (DTG). The detection of hysteresis and the determination of the loading at ambient conditions are challenging.

For open systems another possibility is to use a well-defined humidified carrier gas (e.g. Setaram WetSys) which flows around the sample. To prevent condensation, the transfer line and the measurement cell have to be temperature controlled in an accurate way.

The third possibility is to measure at closed working fluid atmosphere. As shown in (Henninger, Schmidt et al. 2010) measurements with open and closed systems are comparable if using the same reference conditions.

In order to define a common measurement procedure several influencing factors have to be taken into account. An overview is given in Figure 1.

Beside differences in the described measurement method also differences in the measurement processes exist. Some processes are performed at isobaric some processes are performed isothermal. Isothermal measurements in principle allow the determination of the heat of adsorption, by calculation for at least two isotherms or direct measurement within

a simultaneous TG/DSC. In addition, especially with regard to the Dubinin transformation, the temperature independency can be verified.



Figure 1: Influencing factors for thermal analysis of the adsorption characteristics

Isobaric measurement can in principle be performed in a broader temperature range therefore covering a larger range of the adsorption potential $A = RT \ln p/p_0$. Furthermore as the real cycle (ideally) consists of two isobaric phases of desorption and adsorption at condenser and evaporator pressure, the isothermally measured data is not directly adoptable to the operating device.

2.2 Proposed procedure

All of the described aspects need to be regarded when it comes to the definition of a universal valid measurement procedure. As a result of the consideration a common method for determination of water adsorption characteristics with focus on adsorption heat pumps and chillers has been developed (Henninger, Freni et al. 2011). The procedure consists of a pre-treatment of the sample under continuous evacuation (vacuum level: $1e^{-4}$ kPa). The optimal sample pre-treatment temperature should be selected according to the following classification.

- Strongly hydrophilic zeolites (4A, 13X): pre-treatment $T=300^{\circ}\text{C}$.
- Hydrophilic aluminosilicates (NaY): pre-treatment $T=200^{\circ}\text{C}$
- Hydrophobic aluminosilicates (silicalites, ZSM5): $T=150^{\circ}\text{C}$
- Aluminophosphates (AIPO, SAPO): $T=150^{\circ}\text{C}$
- Others (silica gels, activated carbons): $T=150^{\circ}\text{C}$

The sample is heated starting from ambient conditions with a heating rate of 1 K/min followed by an isothermal drying step for another 8 hours. In the following step, isobar measurement at a water vapour pressure of 1.2 and 5.6 kPa takes place.

The selection of the two pressure levels is motivated with respect to the possible applications in TDHP systems. The pressure level of 1.2 kPa corresponds to an evaporation temperature of 10°C , which marks a useful temperature level for cooling applications. The second pressure level of 5.6 kPa corresponds to 35°C which either marks the temperature where heat can be rejected (cooling application) or can be used for low temperature heating (heat pumping application). Figure 2 shows the pressure and temperature profile for the measurement procedure. For each pressure level the sample temperature is varied in 5 or 10 K steps between 150°C and 40°C (for 5.6 kPa) or 20°C (for 1.2 kPa) respectively. In addition at least one adsorption and desorption measurements should be performed in order to detect possible hysteresis effects.

For other refrigerants different pressure steps need to be performed but with properties similar to the described procedure.

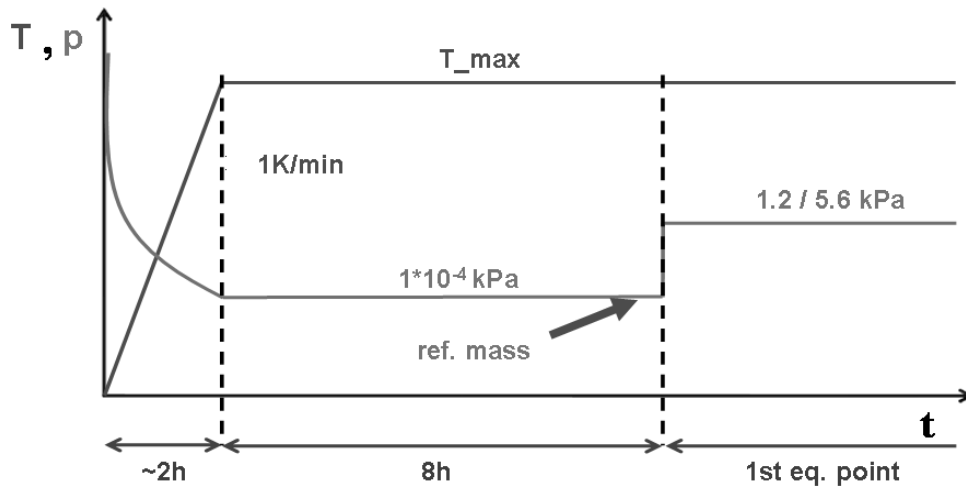


Figure 2: Proposed measurement procedure, including sample pre-treatment and the first isobaric step

The proposed procedure is still under discussion and not yet a standard. This requires further measurements.

3 ADSORPTION HEAT EXCHANGERS

Intensification of the heat transfer quality in adsorbers is a key-factor for development of dynamically efficient adsorption refrigeration and heat pump systems. Several concepts are proposed in literature to contact the adsorbent with the heat exchanger. These concepts can be essentially classified in granular (loose grains) configuration, mechanically consolidated layers and coated heat exchangers HEXs (see Figure 3).

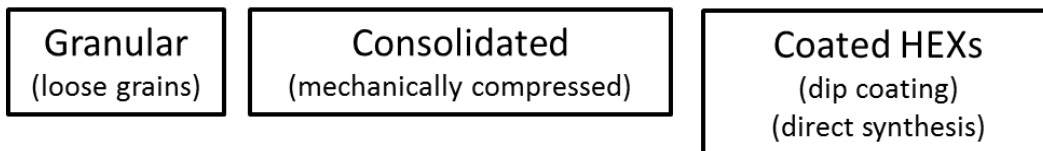


Figure 3: Different adsorber concepts

Consolidation of the adsorbent material is usually achieved by mechanically compressing the adsorbent powder into a mould. Addition of highly conductive compounds (e.g. expanded graphite) is often adopted to further enhance the thermal conductivity of the adsorbent layer (Wang, Metcalf et al. 2012), (Critoph and Metcalf 2011), (Wang, Metcalf et al. 2012), (Kummer, Földner et al. 2012).

The compacted layer may be highly dense (low gas permeability) and rather thick (large pressure drops), so that this solution appears especially suitable for activated carbon/ammonia systems, where the high ammonia pressure allows to avoid limitations in the process rate (Wang, Tamainot-Telto et al. 2011).

Differently, the concept of coating the heat exchanger surface with a thin layer of active material appears appropriate for low-pressure zeolite (or silica gel)/water systems. In this case, heat transfer rate is increased by improving the thermal contact between active material and heat exchanger, rather than the thermal conductivity of the adsorbent layer itself. Proper selection of the coating density and thickness can prevent high mass transfer resistances (Dawoud 2010), (Dawoud, Höfle et al. 2010).

Many coating methods have been reported in literature, including in-situ zeolite crystallization (Bonaccorsi, Freni et al. 2006; Bauer, Herrmann et al. 2009; Schnabel, Tatlier et al. 2010), (Freni, Bonaccorsi et al. 2006), (Freni, Bonaccorsi et al. 2009; Jeremias, Henninger et al. 2012), adhesive coating (Dawoud, Vedder et al. 2007), (Bonaccorsi, Bruzzaniti et al. 2012) and dip coating process (Freni, Russo et al. 2007; Okamoto 2010; Kummer, Földner et al. 2012). Direct accretion of zeolite crystals of the metal surface is a very interesting concept, as the resulting thermal contact is expected to be nearly perfect. However, in order to reach an acceptable zeolite layer thickness (> 0.1 mm), multiple depositions are necessary.

Hydrothermal synthesis of zeolite can be a quite complex and expensive process, especially for the SAPO family, which requires a long treatment in autoclave under high pressure and temperature. Moreover, one has to consider possible problems of different thermal expansion between the zeolite layer and the metal substrate due to the high temperature reached during the treatment (up to 550°C for SAPOs).

The dip-coating method represents an alternate way to deposit a thin layer of adsorbent on the heat exchanger surface. According to the techniques reported in literature, the metal substrate is immersed into a liquid solution made of active powder and an organic (e.g. resins) or inorganic substance (Aluminium hydroxide clays, etc.), acting as a binder. A final thermal treatment is applied to remove the excess solvent, so obtaining a compact adsorbent layer. Advantage of this method is the possibility to easily vary the coating thickness in the range $0.1 - 1$ mm by, for example, controlling the viscosity of the liquid solution and the dipping velocity. Experimental studies on full-scale coated adsorbers returned encouraging results, especially in terms of adsorption cycle time reduced down to a few minutes (Dawoud 2012), that can be translated to an elevated specific power.

However, the presence of binder reduces the overall sorption ability of the resulting coating and - in case of organic compounds - production of volatile substances is possible during operation of the adsorber, so altering the system pressure.

The most simple (and cheap) adsorber concept consists of the embedding of adsorbent grains between the fins of the heat exchanger. It is self-evident that the adsorbent granules present intrinsically poor heat transfer properties. However, recent experimental data demonstrated that a granular adsorber can provide acceptable performance when small-size grains (< 0.5 mm) and extended surfaces heat exchangers (> 1000 m^2/m^3) are employed (Grisel, Smeding et al. 2010; Sapienza, Santamaria et al. 2011).

Figure 4 illustrates the performance enhancement of the adsorber heat exchanger in several development steps as presented in (Schossig, Witte et al. 2011). The left-hand photo shows an automotive cooling unit used to create a packed bed adsorber where the sorbent is prevented from falling out at the bottom by a wire mesh, and a brush helps to fill in the sorbent very closely. Although the volume specific density (sorbent per volume) is very high in this case, the thermal connection between the sorbent and the heat exchanger fin is fairly poor, hindering the released heat (arising by adsorption of the water molecules on the adsorber) to flow from the granules to the heat transfer fluid. The reason is that there exists at most two point contacts per sphere only between the sorbent (sphere) and the heat exchanger fin (the fin gap is greater than sphere's diameter) and the remaining surface area of the sphere has to face a higher thermal resistance (air gap) while transporting the released heat to the fins. To overcome this, for example a binder based dip coating of the lamella heat exchanger can be applied as next step (middle). Here, a uniform connection is realised all over the heat exchanger and only small air gaps (if any) exist between sorbent and fin. Finally, a direct crystallisation connection, as can be seen in the right hand photograph (150 times enlarged), can be done on top of a metallic short fibre structure (sinter-fused structure), giving an enormous increase of surface area (3-D) (see e.g. (Földner, Schnabel et al. 2011)). This combination provides both good thermal conductivity in

the metal and an excellent sorption-material-to-metal-mass ratio (Schossig, Witte et al. 2011). Numerical values for the increase of surface area are available in (Füldner, Schnabel et al. 2011) for example.

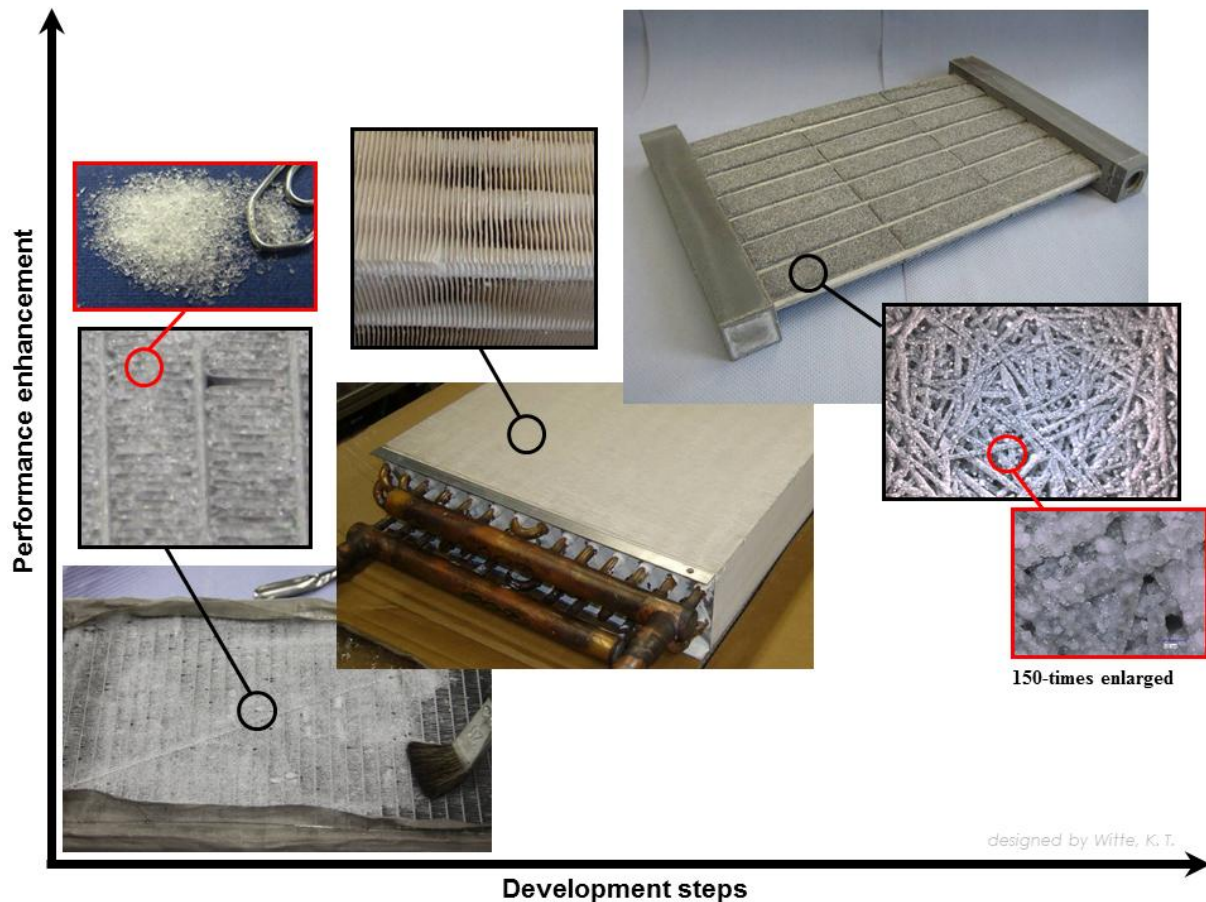


Figure 4: Different evolution steps of the performance enhancement in adsorber development: Packed-bed heat exchanger (left), binder based dip-coated lamella heat exchanger (middle) and direct crystallisation connection of the sorbent (right) on 3D-metal structure heat exchanger. Source (Schossig, Witte et al. 2011)

To evaluate the performance of the coating on the adsorption heat exchanger adequate measurements are required. Such measurement methods are described in (Freni, Santamaria et al. 2012), (Frazzica, Füldner et al. 2012), (Sapienza, Glaznev et al. 2011) and (Sapienza, Santamaria et al. 2011). A comparison between different evaluation and measurement procedures is performed in (Wittstadt, Sapienza et al. 2011).

Since sufficient methods for measurements are available, the design of adsorption heat exchangers can be optimized. An optimization is performed in (Füldner, Laurenz et al. 2012).

4 EVAPORATORS

The evaporator is the heat exchanger that connects the chilled water circuit to the refrigerant. Three major principles exist in the design of the evaporator heat exchangers: falling film, pool boiling and dry expansion evaporators. In falling film evaporators drops flow around the tubes of the heat exchanger tubes and evaporate. In pool boiling the heat exchanger tube bundle is embedded in a pool of refrigerant. In dry expansion evaporators the refrigerant flows in a two phase state through a tube which is heated from the outside. For water as refrigerant only

pool boiling and falling film evaporation is applied since dry expansion evaporation is subjected to higher pressure drops.

Figure 5 shows a tube bundle immersed into a pool as well as a corresponding cross sectional view for pool boiling and falling films. Left hand describes the pool boiling case at a low filling level – typically the tubes are entirely immersed - where the refrigerant (blue) is supplied from the bottom. In contrast the falling film concept is explained via the illustration on the right hand.

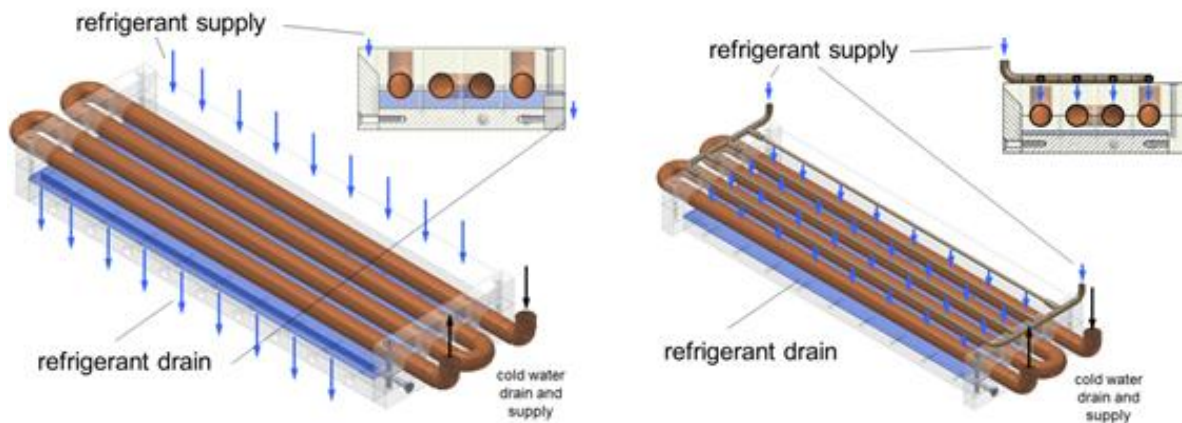


Figure 5: Comparison of pool boiling at a low filling level (left) versus falling film evaporation (right) as investigated in the SORCOOL project at Fraunhofer ISE (see e.g. (Schnabel, Witte et al. 2011))

Improving the heat transfer characteristics of evaporator heat exchangers mainly focuses on two aspects: increasing the heat transfer coefficient between the chilled water circuit and the heat exchanger tubes and increasing the heat transfer coefficient between the heat exchanger and the refrigerant.

For the heat transfer between chilled water and tube wall two options are well known:

- increasing the volume flow inside the tube
- Inserting turbulent flow generators.

Both methods increase the pressure drop within the tubes. Therefore an optimum has to be found between additional electricity consumption and compact design.

Observing the resistances outside the tube several options are available:

- reaching the boiling regime of nucleate boiling
- Lower the thickness of the water layer in the region of convective boiling.

Figure 6 left side shows a high performance 3D-metal structure, manufactured by Fraunhofer IFAM, where the initiation of nucleate boiling has been observed at wall superheats of already 8 K at a pressure of 10 mbar. Tube wetting is reached through capillary surface structuring where water moves upwards automatically. The cut-out on the right side shows high performance evaporation tubes able to make use of this capillary-assisted-evaporation. Additionally, these pictures show turbulent flow generators realised through internal fins (top) and a tube deformation (bottom). Both combinations result in a high tube performance. Thus, performance enhancements of up to factor 14 could have been observed in comparison to a plain reference tube. More information about the evaporator developments for TDHPs can be found in (Schnabel and Weigand 2007), (Schnabel, Scherr et al. 2008), (Witte, Schnabel et al. 2009), (Schnabel, Witte et al. 2010), (Schnabel, Witte et al. 2011), (Witte, Dammel et al. 2011). Measurements of plate evaporators can be found in (Clause, Leprieur et al. 2011).

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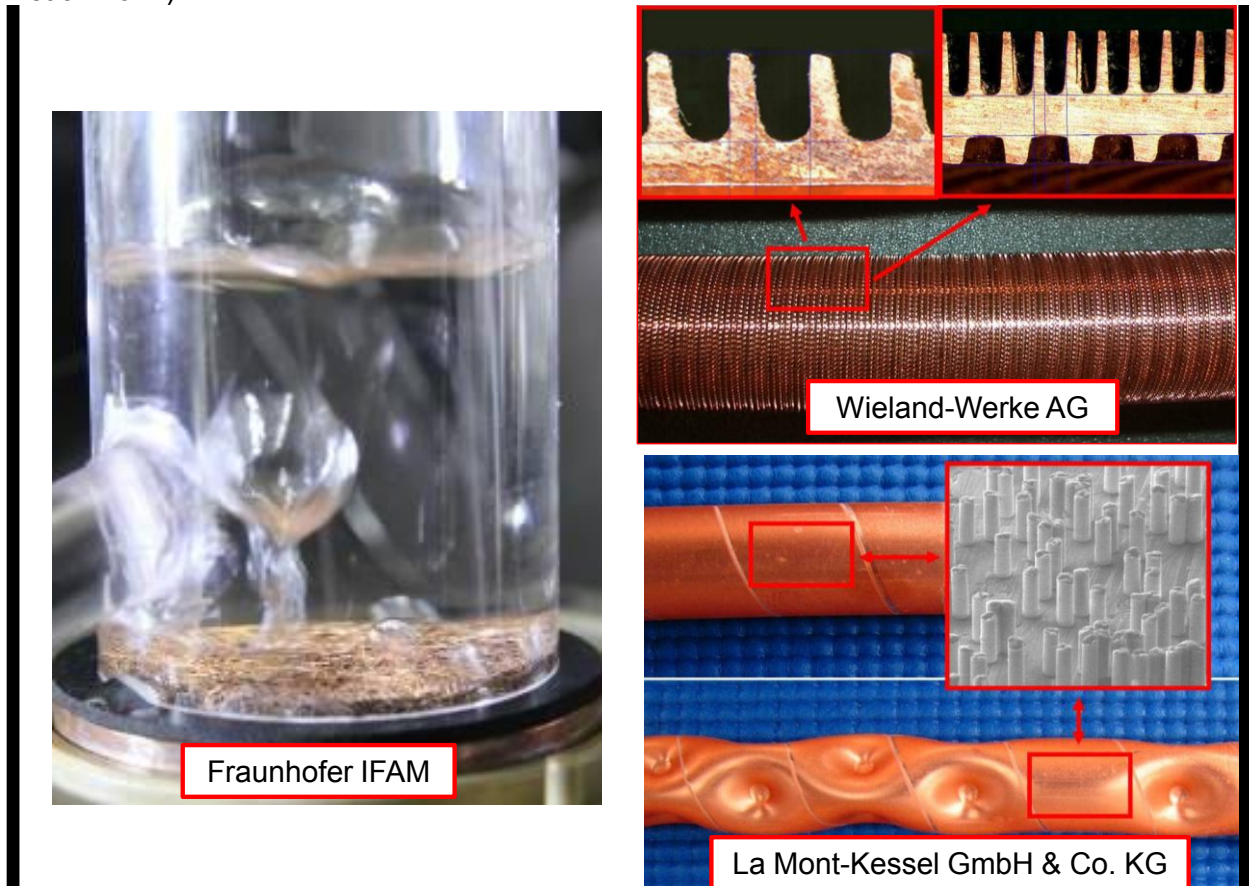


Figure 6: Left: 3D-metal structure manufactured by Fraunhofer Institute for Manufacturing Technology and Advanced Materials (IFAM); top right: high performance evaporation tubes from the manufacturer Wieland-Werke AG; bottom right: So-called Industrial Power Tubes ip tube® from the manufacturer La Mont-Kessel GmbH & Co. KG with a surface treatment by MiCryon Technik GmbH

5 SUMMARY

Current developments on the different components and characterization of adsorption type thermally driven heat pumps were reviewed.

With regard to the basic working pair characteristics, a common measurement procedure for adsorption-refrigerant working pairs is proposed. In addition the activities of different research groups regarding adsorption characteristics, adsorption heat exchanger and evaporator development are summarized.

The proposed procedure for adsorbent characterization covers all measurement aspects, ranging from the pre-treatment and measurement up to the analysis by different thermodynamic models. The method offers the potential to be developed to an international specification or standard.

The current developments on sorption materials, coatings of these materials on supports and new heat exchanger concepts lead to a new generation of adsorbent heat exchangers. However, further improvements are possible regarding the heat capacity and heat and mass

transfer. Due to the cyclic process, a reduction of the heat capacity of the adsorption heat exchanger is beneficial. Increasing the heat and mass transfer ratio, also leads to more efficient TDHPs, as the cycling time can be reduced. However, an optimized sorbent-mass-to-metal-mass ratio has to be chosen regarding the application boundary conditions.

Like in the case for the adsorption material a common measurement procedure for the adsorption heat exchanger is required and under current development.

The improvement of the heat transfer of the evaporator heat exchanger is another important part in order to enhance the overall machine output. Several new developments including different surface structured heat exchangers were reported. The next development step will be the transfer of these new surface structures to a feasible technical scale production. Though, heat exchangers with higher heat transfer coefficients are in direct competition with simply larger heat exchangers. However, the new highly porous surface structures may show a beneficial reduction on the needed metal masses combined with a compact design for a given evaporation power output.

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