

Group: Karlsruhe Liquid metal Laboratory (KALLA)

## **Liquid metal technology research at Karlsruhe Liquid Metal Laboratory 2020 – Fueling sustainable energy and process technology by innovative approaches**

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### **Introduction**

Research and development activities at the Karlsruhe Liquid Metal Laboratory (KALLA) in 2020 can be categorized in three pillars:

- Thermal fluid dynamics of liquid metals
- Liquid metal based heat storage
- Liquid metal based process technology

In all three areas, experimental and modelling approaches are combined, mutually benefitting from each other. All three areas do also benefit from the long lasting liquid metal engineering experience at KALLA and contribute to its progress at the same time. This holds for the design of the loop facilities, reactor setups, instrumentation, components as well as for process control and data acquisition systems. They all define the state of the art in liquid metal technology and enable innovative applications. The latter range from liquid metal based high temperature thermal storage up to highly innovative process routes for e.g. the production of energy carriers.

High temperature heat storage is expected to be an enabling technology for post-fossil heat supply to high temperature thermal process technologies, based on fluctuating sources or waste heat. In terms of high temperature processes itself, liquid metals become more and more visible as heat transfer or even reaction media in highly innovative process routes like direct dehydrogenation of methanol and direct thermal decomposition of methane. While the

first can be part of a process chain to produce sustainable liquid energy carriers, the second can be used as hydrogen production technology or as part of innovative pathways towards utilizing carbon dioxide as a raw material for the production of e.g. carbon black. All of the mentioned topics fit together in a picture of enabling technologies and new ideas for the energy transition with its challenges not only for the energy sector, but for chemistry, transport and many others.

In terms of serving fundamental research, a refurbishment of the GALINKA-loop has been accomplished, that will facilitate precise measurements of the heat transfer behaviour in non-homogeneously heated tubes, delivering both – correlations as well as validation data for sophisticated simulation studies and turbulence model development. A second important experimental topic with direct relevance for simulation tool development, validation and safety assessment of innovative nuclear research installations is the investigation of local blockages and deformations in liquid metal cooled rod bundles. Here we continue the traditional combination of generic investigations with application relevant parameter ranges, resulting in unique data sets that are acknowledged worldwide in nuclear safety research.

We are grateful that - despite the difficulties of the pandemic situation - we could continue the strong partnerships with researchers across Europe and worldwide, and even establish new partnerships with internationally leading companies in the fields of liquid metal and process technology. Please see brief descriptions

of all mentioned activities on the following pages and do not hesitate to contact us, if you see an opportunity to develop creative ideas with the KALLA team in future.

## Thermal Fluid Dynamics of Liquid Metals

### GALINKA – Gallium-Indium-Tin loop Karlsruhe

One of the most traditional installations in KALLA, the liquid metal test loop GALINKA, has been completely renewed in order to realize high-precision liquid metal thermohydraulic experiments at room temperature. The used eutectic composition of gallium, indium and tin has a melting point of 10.5°C. The loop is shown on the left-hand side of figure 1. It hosts a variety of instrumentation to determine mass and volume flow rate, temperatures and pressures at several places in the loop. Experiments with heat loads up to 20 kW, which can be cooled back with a cooling water circuit and a liquid metal volume flow rate up to 1.5 m<sup>3</sup>/h can be realized. Due to the low operating temperature and the low installation space of the loop compared to other liquid metal loops, new experiments can be realized easily and cost effective.

In a current project supported by the German Research Foundation (DFG) (WE 4672/4-1), the influence of an asymmetric thermal boundary condition on the heat transfer in a turbulent liquid metal pipe flow is under investigation. This type of boundary condition can be found in concentrating solar power plants [1, 2]. For this purpose, a test section has been developed to realize either symmetric or asymmetric thermal boundary conditions. Pacio et al. [3] already showed, that even for a symmetric thermal boundary condition high-precision thermohydraulic experiments would be beneficial. Prior to the liquid metal experiments, the new test section has been validated using water at different Prandtl-Numbers and the results were compared to Gnielinski's correlation for heat transfer in turbulent pipe flows [4] showing a

good conformity. An according parity plot can be seen figure 1.

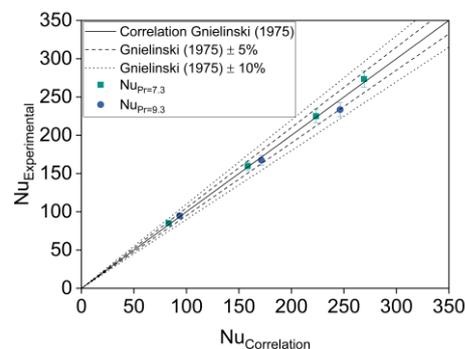
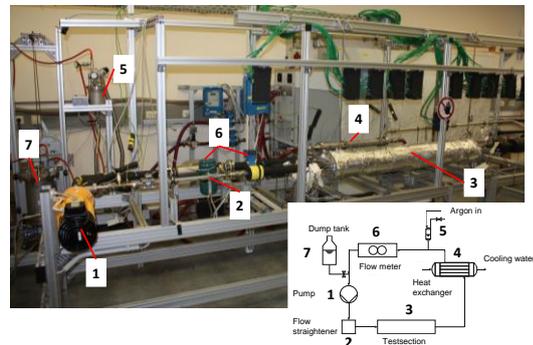


Figure 1: GALINKA loop with all essential components (top), validation of testsection using water experiments and Gnielinski's correlation for heat transfer in turbulent pipe flows (bottom).

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*Fuel assembly experiments in Heavy liquid metal flows*

For the safe and reliable operation of a fuel assembly in the reactor core, the knowledge of the heat transfer to the coolant is essential. Moreover, during the lifecycle of the assembly its geometry can be deformed by swelling, creeping and mechanical defects or blocked by debris. As a consequence, locally reduced cooling and hot spots are expected.

A set of two different experiments and accompanying CFD simulation is planned in support of safety studies for heavy liquid metal cooled fast reactor systems in the EU-Projects PATRICIA and PASCAL. In a first study, the effect of a well-defined porous blockage in a wire spaced 19-pin rod bundle will be investigated. Following the EU-Project MAXSIMA, where a total blockage has been investigated, the effect of a more realistic sintered blockage with well defined porosity will be investigated in PATRICIA. Detailed instrumentation will give insights about local hot spots as well as recirculation patterns which will be used for the validation of numerical models in house by the Framatome Professional School (FPS) Group of the ITES and external partners in the framework of the EU-project. In a second study, the effect of deformations in a rod bundle on the local flow and temperature field will be studied. In the framework of EU-Project PASCAL, a water rod bundle experiment will be set up at the Karman Institute for Fluid Dynamics (VKI), Brussels with design and numerical support by KALLA.

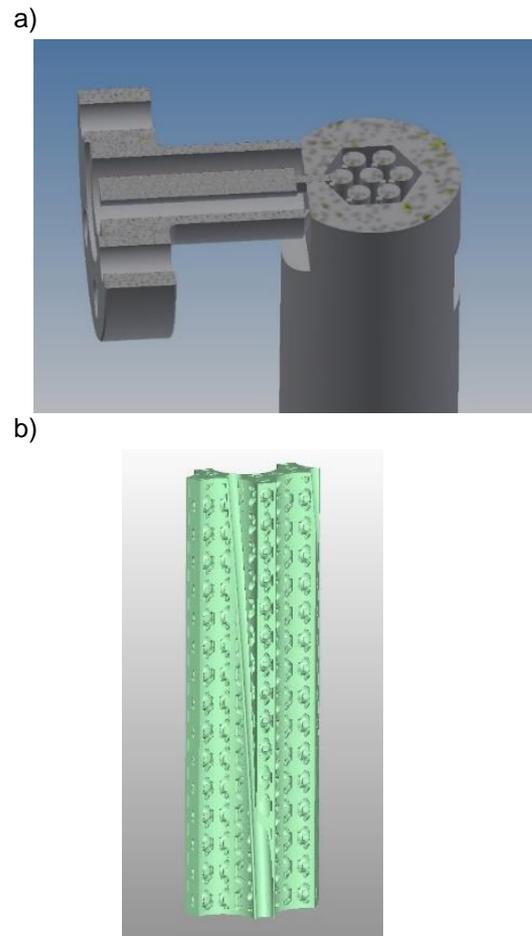


Figure 2: a) Design detail for the planned rod bundle deformation experiment in EU-Project PATRICIA b) Design detail for the planned rod bundle blockage with well-defined porosity in EU-Project PATRICIA



Figure 3: Complete blockage of two sub channels for the experiments in EU-Project MAXSIMA, wires are part of the temperature sensor instrumentation within the blockages and the surrounding rod walls.

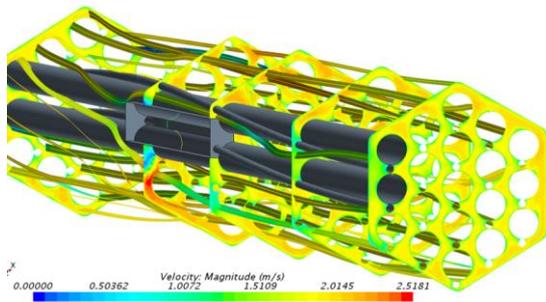


Figure 4: Velocity magnitude plotted on selected planes near the blockage and the streamlines colored with velocity magnitude for the blockage experiments in MAXSIMA<sup>1</sup>.

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## Liquid Metal Based Heat Storage

Liquid metals are tested for the use in thermal energy storage systems at Karlsruhe Liquid Metal Laboratory (KALLA), as they facilitate highly efficient heat transfer in a wide temperature range. A packed-bed storage system with filler material was proposed as a promising storage system in previous theoretical works at KALLA [1,2]. Starting in 2020, experimental tests are carried out, following a series of subsequent experiments with increasing complexity, size and technology readiness level during the upcoming years. First, the operability of a liquid metal thermal energy storage with a packed-bed has been successfully shown in the lab-scale experiment VESPA [5] with spherical ceramic particles as filler material and lead-bismuth as the heat transfer fluid [3,4]. Figure 5 shows a picture of the open storage container filled with zirconium silicate particles.

The material of the filler material has been selected in cooperation with the group of Prof. Müller and Dr. Weisenburger at the Institute for Pulsed Power and Microwave Technology (IHM) at KIT by storing various candidates (ceramics, glasses and natural stones) in lead-bismuth at very high temperatures in order to examine their corrosion behaviour via optical and electron microscopy and other advanced characterization methods. Important lessons regarding filling, draining, cyclic operation, measuring of temperature and flow have been learned for the planned next experiment, featuring a demonstrator with 100 kWh thermal energy storage capacity that is to be built and tested at KALLA.



Figure 5: Packed-bed thermocline storage in the pre-experiment VESPA at KALLA (Photo: F. Müller-Trefzer)

Liquid metals are especially suitable for high temperature applications, as they are liquid up to temperatures of more than 1000°C. However, their corrosive nature requires technical solutions regarding the structural materials that can be used, especially at temperatures beyond 600°C. In this regard, a BMWi-project together with the German Aerospace Center (DRL) and the industry partner KSB, an international producer of pumps and valves, is starting in 2021 regarding testing of pumps and valves at temperatures up to 700°C [6], again also including the colleagues from the group of Prof. Müller and Dr. Weisenburger at IHM.

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### **Liquid Metal Based Process Technology**

#### *DECAGAS<sup>LM</sup> - Decarbonisation of nature gas in liquid metal*

Hydrogen technology is seen as a key to the success of the energy transition. In June 2020, the German government adopted the national hydrogen strategy [1]. These extensive measures and investment packages are intended to establish hydrogen as a predominant energy carrier in the foreseeable future.

In recent years, intensive work has been carried out at KALLA on a continuously operable process for direct thermal methane pyrolysis as a viable process to produce hydrogen and solid carbon from methane and – perspectively – biogas and natural gas. In the process, the extensive experience with liquid metals, established at KALLA, could be transferred into successful lab-scale experiments, which to date have repeatedly proven the possibility to reach high methane conversion rates and their dependency on operation parameters like temperature [2, 3].

Together with the industrial partner Wintershall Dea [4], the technique is now being further developed for the use on an industrial scale. The basis is the pyrolysis of methane in a liquid metal bubble column reactor (see Fig. 6). In this process, liquid tin is flown through by gaseous methane at 800 °C - 1200 °C. Due to the temperature conditions, the methane is split into two components, solid powdered carbon and gaseous hydrogen. Since the solid carbon has a much lower density than the liquid tin, it accumulates on the surface of the liquid metal and can be removed from the reactor. The gas phase, containing a mixture of hydrogen and unreacted methane, will be further processed

according to the requirements of the final application. Unreacted methane and other hydrocarbons can be fed back into the reactor.



Figure 6: Graphical illustration of methane pyrolysis using bubble column reactor. (Infographic: Leon Kühner, KIT) [4]

#### References:

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#### NAMOSYN - Sustainable mobility with synthetic fuels

The BMWi project NAMOSYN (Nachhaltige Mobilität mit synthetischen Kraftstoffen) was created and started in 2019 [1]. Its main aim is to develop and test synthetic fuels that can be

produced and used sustainably. One promising possibility is to use formaldehyde as an intermediate to synthesise, e.g. OME, an alternative to diesel. Moreover, it is also by far one of the most commonly produced chemicals in the industry. Nevertheless, a stoichiometric amount of water is formed in the current methods of CH<sub>2</sub>O production and needs to be separated, which makes these processes costly.

In contrast, the direct dehydrogenation of methanol with sodium vapour as catalyst may allow to avoid the energy-consuming water separation process and to obtain highly concentrated formaldehyde [2]. Despite these potential advantages, there is no industrially mature method for the production of anhydrous formaldehyde yet. The MEDENA set-up is being constructed at KALLA in order to prove or develop according process concepts [3]. A number of parameters, such as the amount of sodium, the mole fraction of methanol and temperature, play a key role in this type of reaction and will be investigated in detail. The MEDENA set-up aims to demonstrate that vaporised elemental sodium homogeneously catalyses the dehydrogenation of methanol to anhydrous formaldehyde with high activity and selectivity.



Figure 7: MEDENA set-up for the dehydrogenation of methanol to anhydrous formaldehyde at KALLA (Photo: M. Kamienowska)

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### NECOC – Negative Emissions by Separation of atmospheric CO<sub>2</sub> into technically usable Carbon Black

The BMWi funded project NECOC (Negative Emissions by Separation of atmospheric CO<sub>2</sub> into economically usable Carbon Black) focuses on greenhouse gas reduction and circular economy based on CO<sub>2</sub>. In collaboration with the two start-ups Climeworks Germany GmbH and INERATEC GmbH a group of researchers from the Karlsruhe Liquid Metal Laboratory (KALLA) and the Institute of Thermal Process Engineering (TVT) is building an innovative demonstration plant [1]. The objective of the project is to combine three process steps to transform atmospheric CO<sub>2</sub> into economically usable, solid carbon.

Atmospheric CO<sub>2</sub> is separated from the air via a direct air capture (DAC) facility developed by Climeworks. The second process step, operated by INERATEC, is the catalytic methanation of CO<sub>2</sub> using microstructured reactor concepts in order to overcome the challenge of the high thermal stability of the CO<sub>2</sub> molecule. In a third step the thermal dissociation of methane, also known as methane pyrolysis, takes place in a liquid metal bubble column reactor developed at KALLA. The innovative technology using liquid tin as heat transport medium prevents the pyrolysis reactor from clogging as the solid carbon produced by methane pyrolysis rises to the liquid metal surface as a powder due to its lower density compared to tin.

Besides demonstrating the feasibility of the combined process another objective of the project is a closer analysis of the pyrolytic carbon. Therefore, and for the continuous operation of the process, a modification of the pyrolysis reactor allowing continuous carbon recovery on lab scale has been developed and tested during the last months. At the same time construction works for the demonstration plant started several weeks ago after detailed planning. In addition to the BMWi funding, an equipment application was successful at KIT within the framework of the Excellence Initiative (KIT future fields) and as a result a very well equipped gas chromatograph could be procured. This will make it possible to identify by-products in the product gas stream across a broad spectrum.

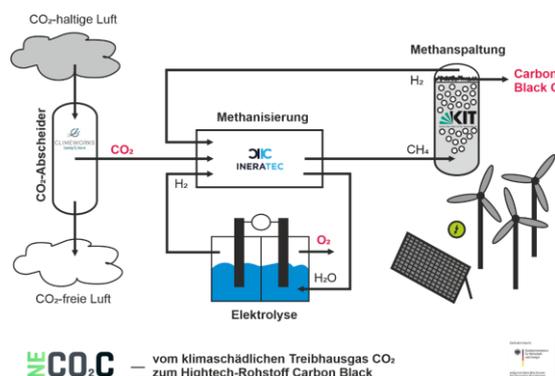


Figure 8: Schematic of the process for converting the greenhouse gas CO<sub>2</sub> into technically usable carbon black, which is currently being set up at KIT in a plant

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