EUROPEAN ROADMAP OF PROCESS INTENSIFICATION

- TECHNOLOGY REPORT -

<u>TECHNOLOGY:</u>
Heterogeneously Catalyzed Solid Foam Reactors

<u>TECHNOLOGY CODE</u>:

<u>AUTHOR:</u> C.P. Stemmet, J.C. Schouten, TU/e

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1. Technology

1.1 Description of technology / working principle

Solid foam catalytic reactors use a solid foam structure (e.g. sponge, but not flexible) as the support for depositing catalyst for the purpose of reacting gas and / or liquid phase reactants. The solid foams may be seen as the inverse of a packed bed of non-porous particles. These materials combine high voidage suitable for low pressure drop application and high geometric surface area (for generation of mass transfer area) and high BET surface area (for depositing the catalyst).

The solid foams, as shown in Figure 1, are available in metals (Al, Ni, Cu, etc.), ceramics (Mullite, TiO_2, SiC, etc.) and carbon (known as reticulated vitreous carbon, RVC). They are characterized usually by an average pore size of the somewhat uniform cells making up the structure; commonly referred to the pores per linear inch or ppi number. The pore size and solid holdup can be varied to achieve a suitable material in terms of the pressure drop and available area (geometric).

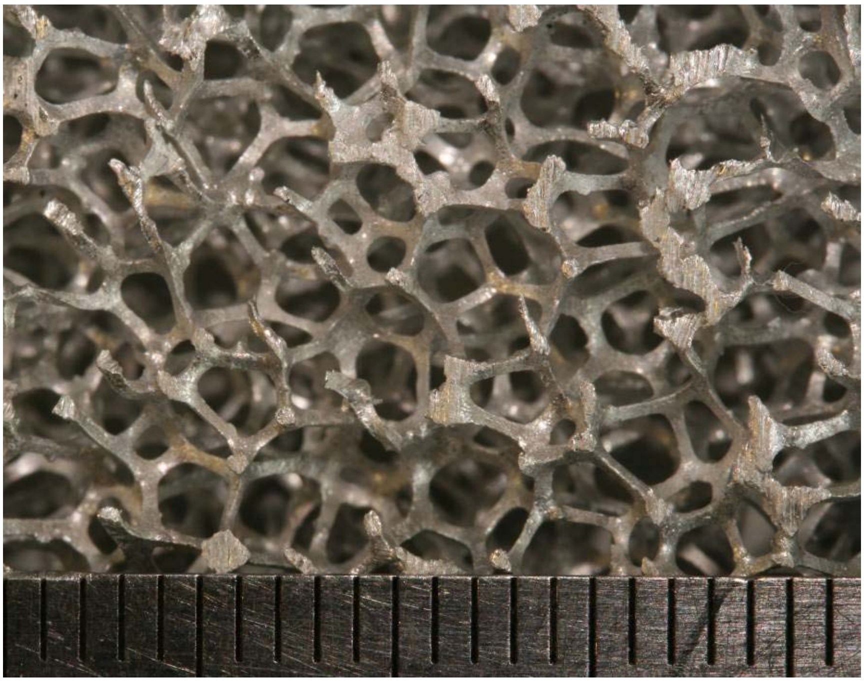

Figure 1: Detail of solid (Aluminum) foam structure, 10 ppi. The graduated markings below are in mm.

Gas and/or liquid flows through the solid foam material as in packed bed applications and reactants are converted to products using suitable catalysts deposited on the surface of the solid foam by standard techniques (wash-coating, ion-exchange, etc). Single-phase applications i.e. where the flowing phase and the solid foam are in contact (gas-solid or liquid-solid mass transfer) and multi-phase contact of all three phases (gas-liquid-solid mass transfer) are proposed. Heat transfer applications are proposed due to the high surface area and high heat conduction of metal foams. Carbon foams are also used as electrodes in electrochemical applications or as catalyst supports due to the high level of chemical inertness.

1.2 Types and "versions"

The reactors envisioned to date using these solid foam structures can be divided into the following distinct versions:

Single phase applications:

Heterogeneous chemical reactors
Gas or liquid flows through the reactor and react on the surface of the solid foam packings to form the products. The solid foam packings act as a support for the catalyst (applied using wash-coating, ion-exchange or other techniques typically used for packed beds). These solid foams are used in CO conversion for applications such as Fischer-Tropsh synthesis and exhaust gas treatment see Richardson et al. (2003), Seijger et al. (2000), respectively. Methanol oxidation or the catalytic production of hydrogen from methanol is studied by Pestryakov et al. (2002) and de Wild and Verhaak (2000), respectively. Determining the pressure drop for solid foam supports for a single phase heterogeneously catalyzed reactor was conducted by various authors, see Innocentini et al. (1999), Montillet et al. (1992), Du Plessis et al. (1994) and Smit and Du Plessis (1999). Each study uses a different model to characterize the foams. The most favorable of these is the model of Fourie and Du Plessis (2002) where they used a tetrakaidecahedra as a representative unit cell (RUC) to predict the geometric surface area for varying ppi number and solid holdup, and thus model the pressure drop using an Ergun-type equation. Pletcher et al. (1991) investigated the fluid-solid mass transfer.

Heat transfer devices (no chemical change)
The gas or liquid flows through the solid foam packing and the high surface area combined with the high thermal conductivity of metal foams is used to dissipate heat, see Boomsma et al. (2003), Lu et al. (1998) and Bhattacharya et al. (2002). Ceramic foam packings are investigated for the use as burners of lean premixed methane in the radiant or blue-flame mode, see Bouma and de Goey (1999), and also investigated as a solar receiver for concentrated solar radiation, see Fend et al. (2004). Ismagilov et al. (2001) have studied the catalytic heat exchanger for the combustion of CH_4.

Electrochemical applications (waste water treatment)
Reticulated vitreous carbon (RVC) as the working electrode for a range of reactions attempting at depositing harmful components Pb(II), Cu, etc. from a solution, see Pletcher et al. (1991), de Leon and Pletcher (1996), Widner et al. (1997), El Deab and Saleh (2003)and Friedrich et al. (2004) for more information.

Photocatalytic applications
Aluminum foam covered in TiO_2 is used in combination with a UV light to achieve photo-degradation (Isopropyl alcohol to CO_2), see Changrani and Raupp (2000). The use of RVC for field emissions cathode for light source applications is studied by Chakhovskoi et al. (2003).

Multi-phase applications:

Heterogeneous chemical reactions (structured packing or catalyst support)
The solid foam structure is used as catalyst support for chemical reaction between reactants in the gas and the liquid phase. The gas and liquid flows through the solid foam packing material and typical configurations found in packed beds (spherical particles or Raschig rings) such as co-current (upflow or downflow) or counter-current flow (flooding limited) can be employed. Carbon nanofibers grown onto the surface of the support can be used to increase the BET surface area. Hydrodynamics such and pressure drop and liquid holdup (dependant on flow configuration) remain favorable

due to the high voidage. Single phase rates of mass transfer, inter-phase gas-liquid mass transfer and the overall activity of the catalyst, are important factors in the design of the overall reactor, as well as the prevailing hydrodynamics. Stemmet et al. (2005) and Stemmet et al. (2007) elucidate on the hydrodynamic parameters and mass transfer when operating the reactor in the counter-current configuration or in the co-current upflow configuration. One of the earliest applications considered was using the foam as a demister.

Electrochemical application
Carbon foam is used as the electrode for hydrogen production from flowing alkaline solution. The bubbles of hydrogen form due to the reaction on the surface and are swept away with the flowing liquid phase as they have deleterious effects on the overall performance and current distributions within the electrode, see El Deab and Saleh (2003).

Rotating packed beds
Cylindrical shapes of solid foam packings are rotated while liquid is fed at the center of the packing material. The liquid flows outward to the edge over the struts making up the solid foam structure. Gas can be also fed at the center to push the liquid as a thin film over the surface. If the rotation of the cylindrical solid foam is increased the liquid flows as a thin film over the solid foam structure. This is done for contacting the gas, liquid and solid for the purpose of a chemical reaction, see Burns et al. (2000) and Guo et al. (2000). Here the liquid holdup (a very important parameter for multiphase applications) is quantified, albeit with high errors. Rao et al. (2004) quantify also the mass transfer coefficients for this type of reactor.

Non-chemical engineering applications:
Composite structures as structural components in cars and housing
Optics (lightweight composite mirrors)
Gas flow control
Cryogenics
Filters (foundry metals)
Semiconductor industry (diffuser disks for fluid flow, plasma stabilizer, condensation of by-products, efficient heat transfer)

1.3 Potency for Process Intensification: possible benefits

Table 1: Documented and expected benefits resulting from technology application

Benefit	Magnitude	Remarks
Energy Savings	Order of magnitude	When the solid foam is used as a catalyst support for chemical reaction part of the energy dissipated is lost through mixing and churning which do not contribute to the overall performance. However, most of the energy is efficiently used to generate gas-liquid mass transfer which ultimately results in higher overall conversion. The order of magnitude in energy saving is achieved by comparing the up-flow reactor to the trickle bed reactors most currently used. Other packings can also be used, but suffer from “dead” volume which ultimately results in lower volumes for gas-liquid flow, higher pressure drop and mass transfer limitations into the solid (diffusion limitations).

Increased yield	No data available	The gas-liquid mass transfer in multiphase reactors is usually the rate determining process. However, for solid foam packings this gas-liquid mass transfer is energy efficient and increases with the liquid velocity through the reactor. The overall reactor (catalyst and open volume) can be engineered for optimum performance.
Catalyst savings	No data available	See above.
Better heat transfer	See single phase heterogeneous experiment	Reaction heat may possibly be added or removed when using a metal foam to avoid hot-shot formation.
Fouling prevention	No data available	Due to the structure of the solid foam the liquids are filtered (see Non-chemical engineering applications) and so prevent fouling of the overall reactor.

1.4 Stage of development

The use of solid foams as a catalyst support material for multiphase reactions is in its infancy as the previous research has focused on using the material for single phase applications. Stemmet et al. (2007) outline some of the hydrodynamic properties and gas-liquid mass transfer coefficients for a co-current upflow reactor. This reactor operated more efficiently than the downflow version operated as a trickle bed with the solid foam as packing material. This efficiency is also evident in using other (more conventional and structured) packing materials, but these suffer from high pressure drop at similar gas and liquid velocities.

2. Applications

2.1 Existing technology (currently used)

The slurry bubble column reactor is used currently for a vast majority of the multiphase heterogeneous catalyzed reactions, many operated in batch mode especially in the pharmaceuticals industry. The stirred tank is also used for this application with the catalyst supplied on the surface of small (70μm) particles to avoid liquid-solid mass transfer. However, these two reactors suffer from low gas-liquid mass transfer and inefficient use of energy (supplied by the gas-phase or by rotation of a stirrer) due to the high level of turbulence dissipated during the mixing of the liquid phase. This can be viewed as wasted energy as gas-liquid mass transfer (usually the rate limiting step in multiphase reactors) is low compared to other reactor designs. The trickle bed packed with spherical particles is used for continuous operation but also has low gas-liquid mass transfer and relies on gravity assisted drainage of the liquid phase over the packing surface. Depending on the temperature and heat of reaction, heating is applied via a healing/cooling jacket.

2.2 Known commercial applications

Thus far no known applications of the technology as described above are in commercial development or use.

Table 2. Industrial-scale applications of the Technology (existing and under realization)

Sector	Company - Process/Product name/type	Short characteristic of application	Production capacity /Plant size	Year of application	Reported effects
None					•

2.3 Known demonstration projects

So far no known application of this technology is being considered.

Table 3. Demonstration projects related to the technology (existing and under realization)

Sector	Who is carrying out the project	Short characteristic of application investigated, including product name/type	Aimed year of application	Reported effects
None				•

2.4 Potential applications discussed in literature

The potential for using solid foams as the catalyst support material for heterogeneous catalyzed multiphase reactions is quite large. Applications such as Fischer-Tropsch synthesis, gas treatment and hydro-desulphurization have been mentioned, but only single phase experiments have been performed, see Twigg and Richardson (2007), Seijger et al. (2000). Most of the research performed for single phase hydrodynamics has been confusing as researchers apply models and dimensionless analysis before clearly reporting on the results obtained. This list however can be extended to expoxidations, alkylations, hydrogenations, oxidations, hydrocrcaking, hydro-treating and continuous pharmaceutical production once important factors such as depositing a sufficient amount of active catalytic material on the surface of the support has been realized. Also applications to be considered are those in the field of reactive distillation where reactions and separation processes occur in the same volume. Here the solid foam can be used as the packing material for the column and certain sections made catalytically active.

3. What are the development and application issues?

3.1 Technology development issues

Table 4. Technology development issues

Issue	Description	How and by whom should be addressed?
Engineering and design concepts for commercial scale reactors	The multiphase reactor studies thus far performed show that this technology is only in its infancy and development of the technology should be addressed. Likely routes are the performing of reactive studies using the solid foam as a support. Possibly in combination with	R&D projects carried out at universities in collaboration with catalyst manufacturers and producers of industrial scale reactors,

	the study of heat transfer. Also reactive distillation concepts could benefit from this technology	possibly with companies who have suitably flexible technologies (e.g. reactive distillation) to assist in the development
Modeling and scale-up technologies	Research in the field of solid foam multiphase reactors has shown that the scale-up of this technology could be easy as the gas-liquid mass transfer (usually the rate determining step in multiphase reactors) is largely affected by the velocity of the flowing phases. Similar to monoliths the mass transfer is affected largely by the thin film of liquid remaining on the surface as a gas bubble passes. The overall process is independent of the pore size of the solid foam and the size of the packing if the gas and liquid move through the solid foam and no by-passing occurs. This could indicate a development in adequate distributor/re-distribution technology.	R&D projects carried out at universities in collaboration with equipment manufacturers and producers of industrial scale reactors
Production of the solid foam	The solid foam structures produced currently are aimed at a variety of applications and the uniformity of the cell structure cannot always be guaranteed. Also for using this material as a catalyst support the material should be of the open-cell type to allow flow through the pores of the material.	Manufacturers of the solid foam materials may aim at improving their production methods giving uniform cells with a given geometric surface area. Also possibly incorporating the catalytic components into the production procedure

3.2 Challenges in developing processes based on the technology

Table 5. Challenges in developing processes based on the technology

Challenge	**Description**	**How and by whom should the challenge be addressed?**
Catalyst deposition	Due to the high voidage (open volume) of the solid foam material the deposition of active material on the solid foam surface is difficult. This is due to a high liquid volume (and high concentration of the active catalyst precursor) required for deposition of the active catalyst	This challenge should be addressed in the R&D projects in the universities with the assistance of catalyst manufacturers
Washcoat technology	The above mentioned problem may partially be solved by application of washcoats or other high surface area components on the surface of the solid foam material potentially decreasing the concentration of catalyst precursor to be used and increasing the overall dispersion (decreasing the average size of the catalytic sites)	This challenge should be addressed in the R&D projects in the universities with the assistance of catalyst manufacturers
Heat transfer	The heat transfer benefits that may be attained when using a multiphase system and the possibility of adding/removing reactions heat to the overall volume of the reactor to avoid “hot-spot” formation	This challenge should be addressed in the R&D projects on engineering & design concepts in collaboration with universities
Cost	The solid foam materials are being used in a wide variety of applications; however, the cost	This challenge should be addressed in R&D

	of the material remains quite high. Adding catalyst to the solid foam is relatively expensive technology and may be incorporated in the production procedure of the solid foam materials	projects on engineering & design in collaboration with the producers of solid foam

4. Where can information be found?

4.1 Key publications

Table 6. Key publications on the technology

Publication	Publication type (research paper/review/book/ report)	Remarks
Smit, G.J.F. and Du Plessis, J.P.,1999, Modeling of non-Newtonian purely viscous flow through isotropic high porosity synthetic foam, Chemical Engineering Science, **54**, pg. 645-654.	Research paper	Review of models used to describe single phase non-Newtonian flow through solid foams.
Pletcher, D, Walsh, F.C. and Millington, J.P., 1991, Reticulated vitreous carbon cathodes for metal ion removal from process streams, Part I: Mass transport studies, Journal of applied electrochemistry, **21**, pg. 659-999.	Research paper	Single phase, fluid-solid mass transfer.
Stemmet, C.P., Jongmans, J.N., van der Schaaf, J., Kuster, B.F.M., Schouten, J.C., 2005, Hydrodynamics of gas-liquid counter-current flow in solid foam packings, Chemical Engineering Science, **60**, pg. 6422-6429.	Research paper	Hydrodynamics (liquid holdup and pressure drop) for two-phase air/water system operated in counter-current flow
Stemmet, C.P., Meeuwse, M, van der Schaaf, J., Kuster, B.F.M., Schouten, J.C., 2007, Gas-liquid mass transfer and axial dispersion in solid foam packings, Chemical Engineering Science, **62**, pg. 5444-5450.	Research paper	Hydrodynamics and gas-liquid mass transfer operated in co-current upflow configuration.
Scheffler, M., Colombo, P., 2005, Cellular ceramics: structure, manufacturing, properties and applications, Wiley-VCH GmbH, Weinheim, Germany, ISBN, 3-527-31320-6.	Book	Section 4.2 gives information of the permeability and section 5.4 on “heterogeneously catalyzed processes with porous cellular ceramic monoliths”. Section 5.11 gives “Other developments”

4.2 Relevant patents and patent holders

Table 7. Relevant patents

Patent	Patent holder	Remarks, including names/types of products targeted by the patent
GB 1097473 (1969)	B.J. Lerner	Gas-liquid contacting device
US 4251239 (1981)	R.A. Clyde, W.B. Crandall	Multi-purpose ceramic element
US 4810685 (1989)	M.V. Twigg, W.M. Senelow, Imperial Chemical Industries PLC, England	Gas phase water gas shift reaction
WO 01/60515 (2001)	Kourtakis, K., Gaffney, A.M., Conoco Inc, Unites states	Synthesis gas production on ceramic foam
US 2003/0116503 A1 (2003)	Y.Wang, Y. Chin, Y. Guo, C.L.Aardahl, T.L. Stewart	Solid foam structure covered in carbon nano-tubes for gas-phase Fischer-Tropsch
DE 10208711 A1 (2003)	BASF, Germany	Ceramic foam with monolithic type channels down the structure
WO 2006/065127 A1 (2006)	Stemmet, C.P., Kuster, B.F.M. van der Schaaf, J., Schouten, J.C., STW	Solid foam as catalyst support structure for multi-phase contacting and reaction

4.3 Institutes/companies working on the technology

Table 8. Institutes and companies working on the technology

Institute/Company	Country	Remarks
Eindhoven University of Technology, Eindhoven (Prof. J.C. Schouten), and University of Twente, Enschede (Prof. L. Lefferts)	The Netherlands	Heterogeneous catalyzed multiphase reaction using the solid foam as catalyst support and growing of carbon nanofibers on the surface
Delft University of Technology (Prof. F. Kapteijn and Prof. J. Moulijn)	The Netherlands	"Structured catalytic packings to exploit a highly active Fischer-Tropsh catalyst"
ETH, Zurich (Prof. D. Poulikakos)	Switzerland	Thermal conductivity of solid foams
Queen's University, Ontario (Prof. B.A. Peppley)	Canada	An air-breathing PEM fuel cell which uses porous carbon foam (RVC) in the cathode side

5. Stakeholders

5.1 Suppliers and developers

The main suppliers for the solid foam materials can be found on the website http://www.metalfoam.net (or http://npl.co.uk), but the main producers of open-celled solid foam have been listed below in the table below:

Table 9. Supplier and developers

Institute/Company	Country	Remarks
Inco Special products	Ontario, Canada	Nickel foams
Porvair advanced materials	Hendersonville, USA	Cu, Ni, Ti, Fe, Pt, Ag and many alloy foams
ERG Aerospace	Oakland, USA	Aluminum, SiC and Carbon foams
Recemat International	Krimpen aan den Ijssel, The Netherlands	Ni, Ni-Cr and Ni-Cr-Al foams
M-Pore	Dresden, Germany	Aluminum foams
Metafoam Technologies	Quebec, Canada	Cu, Ag, Ni, Fe, Ti foams
Mitsubishi Materials	Japan	
Ultramet	Pacoima, USA	W, Re, Ta, Mo, Nb (Cb), and Platinum group metal foams
SAS Solea	Boussens, France	Porous metals
Spectra-Mat, Inc.	Watsonville, USA	Porous refractory metals
Midvale Foundry Products	USA	Ceramic foam filters
Graftech International Ltd.	Parma, USA	Carbon foam
Ecoceramics	Velsen, The Netherlands	Ceramic (mullite) foam
Vesuvius	Champaign, USA	Ceramic foam

5.2 End users

The likely end users of this technology are the users of the vast majority of heterogeneous catalyzed reactors employing reactants in two or more phases. The pharmaceuticals sector could benefit from this technology where continuous production methods could be used. Catalysts like platinum or enzymes could be supported on the solid foam and a series of these reactors could replace batch processes currently used. Petrochemical industries could possibly use the solid foam material instead of the trickle bed reactors currently being used to apply filtering prior to processing where contacting of phases and chemical reaction is performed efficiently. The users of slurry bubble column reactors may also benefit from having the catalyst fixed within the reaction volume avoiding filtering of the slurry to retain catalyst. The liquid in a semi-batch mode as in slurry bubble columns can also be used with the solid foam, due to the open structure (up to 97%).

6. Expert's brief final judgment on the technology

The benefits to the chemical/pharmaceutical/petrochemical industries by using solid foam as the catalyst support for multiphase heterogeneously catalyzed reactions are widespread encompassing energy savings, efficient use of the reaction volume etc. Although the technology is in the developing stages the potential for this technology in terms of energy efficiency is substantial. The R&D challenges opposing the development of this technology can be met with support from the industries supplying the solid foam in collaboration with universities and the envisioned end users. This effort could provide an industrial-scale reactor using this technology within the next 5-10 years. The energy efficiency on the long term will provide the most important incentive for implementing these types of catalyst supports over the existing technologies due to the reduction in energy usage and ultimately the production of CO_2.

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