CAD Technique for Scaling Up Reactors of Microwave-Assisted Chemistry

Andrew O. Holmes[1], Ethan K. Murphy[2] and Vadim V. Yakovlev[1]

[1] Worcester Polytechnic Institute, Worcester, MA, USA
[2] Applied Mathematics, Inc., Gales Ferry, CT, USA

Scaling up efficient reactions in microwave-assisted chemistry to the level of industrial technology is slowed by difficulties in development of large-scale and highly productive reactors. This paper shows that a microwave reactor can be designed to enhance and accelerate chemical reactions with the help of computer optimization that maximizes energy coupling. We outline an approach based on the application of an efficient neural network optimization technique which is used to optimize the geometry of a system designed as an enlargement of an existing reactor. The approach is illustrated by examples of three-parameter optimization guaranteeing at least 90% energy efficiency.

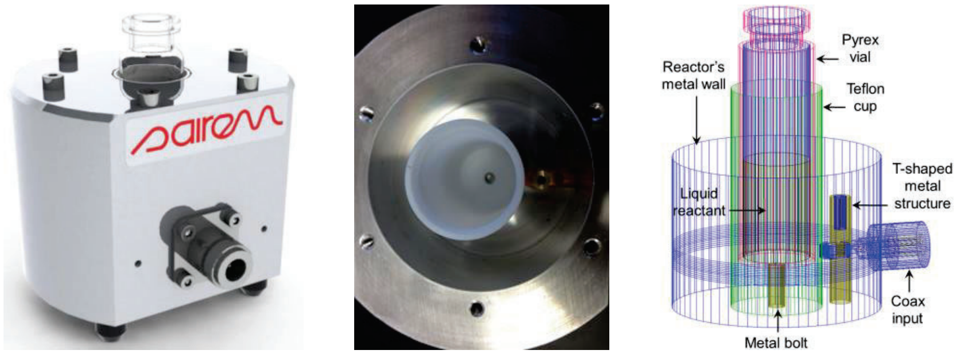
Keywords: Energy efficiency, microwave reactor, neural network optimization.

INTRODUCTION

Microwave-assisted chemistry (MAC) is a promising and quickly growing field utilizing microwaves as the means of enhancing and accelerating reactions in organic/inorganic chemistry, biochemistry, and other disciplines. In comparison with conventional heating methods, microwave (MW) heating provides higher reaction rate, higher chemical yield, lower energy use, and other benefits [1-3]. With special dedicated equipment for MAC now available, particular attention has been recently paid to the development of controllable and reproducible reactions for large-scale production of chemical substances [3, 4].

In order for a MW-assisted reaction to become an industrial technology, there is a need to develop equipment capable of producing new chemical entities on a scale of dozens/hundreds of kilograms. However, increasing the productivity of a process to the level of industrial technology is still considered a challenging problem due to significant difficulties in development of large-scale reactors. This problem is usually addressed via trial-and-error experiments aiming to find correlation between the input parameters of the scaled up MW reactors and the chemical characteristics of the resulting products [5, 6].

This paper shows that laborious scaling up can be made easier by using suitable electromagnetic CAD techniques. We aim to scale up an existing reactor capable of handling liquid reactants of 10 ml volume to larger MW systems designed for processing reactants of 200 ml volume. To improve energy coupling in the larger system, we apply an optimization procedure [5, 7] based on RBF network optimization with CORS sampling. Energy efficiency of the optimal geometries is guaranteed to be at least 90%.



(a) (b) (c)

Figure 1. SAIREM's *MiniFlow 200SS* reactor: general view (a), view of the interior with the upper lid removed (b), and 3D view of the reactor in the developed *QW-3D* model.

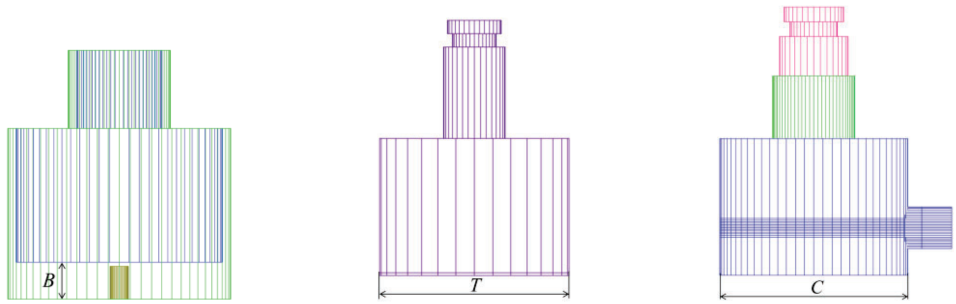
REACTOR AND ITS MODEL

The reactor shown in Figure 1 (a) was produced and put on the market (as *MiniFlow 200SS*) by SAIREM SAS, Neyron, France. The reactor consists of a cylindrical cavity (of i.d. 63 mm) containing a concentrically positioned cylindrical Teflon cup (of o.d. 28 mm) (Figure 1 (b)) that holds a cylindrical Pyrex vial with a reactant inside. The resonator is fed via a coaxial cable by a solid-state generator providing robust control of the frequency of excitation (from 2.43 to 2.47 GHz with a step of 0.1 MHz) and the level of MW power (from 0 to 200 W with a step of 1 W). The reactant in the vial is put into the system through a cylindrical hole in the cavity's lid.

This reactor was reproduced in a parameterized 3D model developed for the full-wave 3D conformal FDTD simulator *QuickWave-3D* (*QW-3D*) ver. 2012; the model was originally reported in [6]. The layout of the reactor's components in the model is shown in Figure 1 (c). Due to the unique opportunity provided by the *MiniFlow 200SS* system to rigorously control frequency and power of the MW output, it was feasible to directly verify performance of the model against corresponding experiments. Comparison of measured and simulated results in [6] has confirmed adequacy of the developed model.

DESIGN OF AN ENLARGED REACTOR

Here we aim to design a MW system similar to *MiniFlow 200SS*, but larger in dimensions and suitable for handling larger loads. While the typical amount of a liquid reactant in a Pyrex vial (Figure 1 (c)) placed in *MiniFlow 200SS* is 10-15 ml, in the enlarged system, we set the goal to process $V = 200$ ml. Since the diameter of the cylindrical vial and the hole in the lid cannot be increased, we consider a modified shape of the vial: instead of a straight cylinder, we use a vessel consisting of two cylindrical portions, as shown in Figure 2 (b). A corresponding two-cylinder shape is also adopted for the Teflon cup surrounding the vessel (Figure 2 (a)). The T-shaped structure used for excitation of the dominant TM mode remains of the same size, but the Teflon cup is no longer required to be in direct contact with the tip of the excitation structure. These changes in the design have been implemented in the *QW-3D* model of the enlarged reactor. In this paper, we employ this model to generate massive amounts of data for subsequent optimization.



(a) (b) (c)

Figure 2. Elements of the enlarged reactor and design variables in optimization: Teflon cup and *B* (a); vessel and *T* (b); reactor cavity and *C* (c).

OPTIMIZATION PROBLEM

Our CAD goal is to find a configuration guaranteeing, for a given process, lowest reflections from the system (and hence highest energy efficiency). We identify in the enlarged reactor 3 geometrical parameters (Figure 3) whose variations make an impact on the frequency characteristic of the reflection coefficient $|S_{11}|(f)$: thickness of the bottom of the Teflon cup (*B*), o.d. of the vessel's low cylinder (*T*), and i.d. of the reactor's cavity (*C*); they are chosen as design variables for optimization. The optimization problem is formulated as follows: for a given reactant, find *B*, *T*, *C* which yield less than E_r% of reflected MW energy (i.e., $|S_{11}| < 0.1\sqrt{E_r}$) in *W*% of the frequency range (f_1, f_2).

We use the RBF neural network optimization with CORS sampling introduced in our works [5, 7], which is capable of quick convergence to the optima due to a reduced number of underlying 3D FDTD analyses (DB). The optimization problem is performed for 200 ml of water, and we apply the constraints $W = 90\%$, $E_r = 9\%$, $(f_1, f_2) = (2.43, 2.47)$ GHz. Since *T* and *C* are not independent, we allow them to be in the intervals:

Problem A: $46 \le T \le 58$ mm; $98 \le C \le 110$ mm; $0 \le B \le 10$ mm
Problem B: $46 \le T \le 62$ mm; $102 \le C \le 118$ mm; $0 \le B \le 10$ mm
Problem C: $46 \le T \le 66$ mm; $106 \le C \le 126$ mm; $0 \le B \le 10$ mm

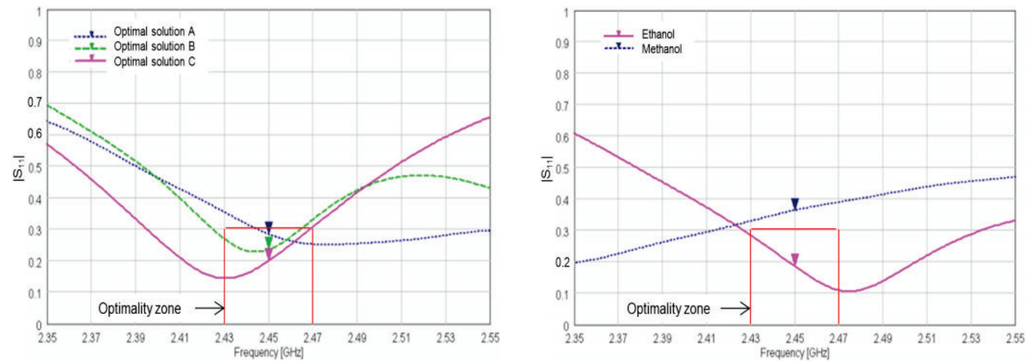
The resulting optimal characteristics of $|S_{11}|(f)$ are shown in Figure 3 (a); corresponding geometrical parameters of the optimized enlarged reactor are:

Water: Solution A: $T = 55.8$ mm; $C = 98.0$ mm; $B = 0$ with $W = 62.5\%$ (DB: 100 pts)
Solution B: $T = 62$ mm; $C = 104.8$ mm; $B = 2.0$ mm with $W = 90\%$ (DB: 57 pts)
Solution C: $T = 50$ mm; $C = 126.0$ mm; $B = 10.0$ mm with $W = 95\%$ (DB: 100 pts)

We also solve Problem B for 200 ml of ethanol and methanol. The best resulting $|S_{11}|(f)$ characteristics are shown in Figure 3 (b); they correspond to the following configurations:

Ethanol: Solution B: $T = 58.5$ mm; $C = 110.0$ mm; $B = 0$ with $W = 100\%$ (DB: 28 pts)
Methanol: Solution B: $T = 50.0$ mm; $C = 103.4$ mm; $B = 10$ and $W = 0\%$ (DB: 100 pts)

It turned out that while the optimization provides an excellent solution within the space of accepted design variables for ethanol, there is no configuration of the enlarged reactor that provides the required level of energy efficiency for methanol.



(a) (b)

Figure 3. Optimization output: reflection coefficient versus frequency for 200 ml of water (a), ethanol and methanol (b) as the reactants of the enlarged reactor.

CONCLUSION

The results of this paper confirm excellent suitability of the neural network optimization [5] for determining a scaled-up configuration of a MAC reactor that possesses desirable energy efficiency. The geometry has been determined from three-parameter optimizations in which the objective function for $|S_{11}|$ is minimized at a small frequency range around 2.45 GHz. We have considered an existing apparatus (*MiniFlow 200SS*); this allows us to experimentally verify the model prior to optimization, and emphasizes the practicality of our approach. The technique works for scenarios with more design variables, more complex configurations, changing media parameters and different reactants. The presented results suggest that CAD of highly productive, large-scale reactors is a useful tool in their design, and that application of the presented optimization procedure may be practical for the expedient design of energy efficient MAC systems.

ACKNOWLEDGEMENT

This research was supported in part by an appointment to the Faculty Research Participation Program at the U.S. Air Force Research Laboratory administered by the Oak Ridge Institute for Science and Education through an interagency agreement between the U.S. Department of Energy and USAFRL. The authors are grateful to SAIREM SAS (Neyron, France) for providing the *MiniFlow 200SS* system for experimentation and modeling.

REFERENCES

[1] P. Lidstrom, J. Tierney, B. Wathey, and J. Westman, *Tetrahedron*, vol. 57, pp. 9225-9283, 2001.
[2] C.O. Kappe and A. Stadler, *Practical Microwave Synthesis for Organic Chemists: Strategies, Instruments, and Protocols*, Wiley-VCH, 2009.
[3] N.E. Leadbeater, Ed., *Microwave Heating as a Tool for Sustainable Chemistry*, CRC Press, 2010.
[4] J.M. Kremsner, A. Stadler, and C.O. Kappe, *Top Curr Chem*, vol. 266, pp. 233-278, 2006.
[5] E.K. Murphy and V.V. Yakovlev, *ACES J.*, vol. 45, no 12, pp. 1108-1117, 2010.
[6] A. Holmes, C. Yang, and V.V. Yakovlev, In: *IEEE MTT-S Intern. Microwave Symp. Dig*. (Seattle, WA, June 2013) (to be published).
[7] E.K. Murphy and V.V. Yakovlev, *Intern. J. of RF & Microwave Computer-Aided Engng*, vol. 21, no 2, pp. 279-287, 2011.