

CAD of Microwave Chemistry Reactors with Energy Efficiency Optimized for Multiple Reactants

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Abstract: Upgrade of successful processes of microwave-assisted organic synthesis to the level of industrial technologies is currently slowed down by difficulties in experimental development of large-scale and high-productive reactors. This paper proposes to address this issue by developing microwave chemistry reactors as microwave systems rather than black-box-type units for chemical reactions. We suggest an approach based on application of a neural network optimization technique to a microwave system in order to improve its coupling (and thus energy efficiency). The RBF network optimization with CORS sampling introduced in our earlier work and capable of exceptionally quick convergence to the optima due to a dramatically reduced number of underlying 3D FDTD analyses is upgraded here to account for additional practically important condition requiring the reactor's optimal design for different reactants. Efficiency of the approach is illustrated by two examples optimizing the geometry of a conventional microwave reactor for 3 different materials: with 1/5 liter reactants, seven-parameter optimization yields the best configurations from only 297/195 FDTD simulations on a regular PC.

Keywords: FDTD simulation, microwave reactor, neural network, optimization, scaling up.

1. Introduction

Microwave (MW)-assisted organic synthesis (MAOS) has recently become a frontline methodology in chemistry programs of pharmaceutical, agrochemical, and biotechnology industries due to the ability of this multidisciplinary technology to significantly speed up chemical reactions [1-3]. With specialized microwave systems now available, particular attention is currently paid to the problem of development of controlled MAOS featuring new reaction routes for organic synthesis resulting in large-scale production of chemical substances [3]. In order for MAOS to become a widely accepted industrial technology, there is a need to develop techniques routinely producing new chemical entities on a scale of dozens/hundreds of kilograms. However, surveying the contemporary literature, one may notice that progress in this direction is fairly slow. While scaling up successful processes of microwave chemistry is commonly acknowledged to be a key issue of the current state of the field, this problem is being addressed via essentially trial-and-error experiments aiming, with no way to measure the temperature and pressure inside the reactant, to catch correlation between the input parameters of the scaled up microwave reactors and the output chemical characteristics of the products [4].

Comprehensive modeling of interactions of the reactants with the electromagnetic field in closed systems appears to be a powerful tool applicable to the microwave chemistry reactors. Specifically, the modeling can be used for determining reflections (i.e., for finding their energy efficiency) as well as spatial distributions of dissipated power in the reactant; application of corresponding computational procedure to microwave systems suitable for MAOS is exemplified e.g., in [5]. There are also multiphysics modeling techniques computing temperature fields in the processed materials by coupling electromagnetic and thermodynamic simulations and taking into account thermal changes of material parameters [6]. Moreover, there are modeling-based procedures optimizing microwave applicators in terms of energy efficiency [7] and synthesizing optimal processes of microwave treatment resulting in the homogenized temperature fields [8]. Being known and used in other microwave power applications, the advanced computational approaches seem to not be utilized in microwave chemistry. Examples of employing computer simulation in design of MAOS reactors as microwave systems are rare (e.g., [9], [10]) and limited to the analysis (rather than optimization or synthesis) of the reactor's operational regimes.

In this paper, we present an original technique for direct computer-aided design (CAD) of microwave reactors of a desirable scale and high energy efficiency for different reactants to be processed there. The approach is based on the radial basis function (RBF) network optimization algorithm described in [11] and featuring (i) an objective function (OF) measuring the bandwidth of the frequency characteristic of the reflection coefficient over a specified frequency range and (ii) constrained optimization response surface (CORS) technique [12] selecting additional sample points in the dynamic training of the network. The algorithm backed by 3D FDTD data is characterized by very quick convergence to the optima and a dramatically reduced number of underlying analyses; due to these features, it has a strong potential for viable optimization of complex microwave systems and/or structures with a large number of design variables.

Here, the technique [11] is upgraded by incorporating an additional practically important condition requiring optimality of the reflection coefficient for multiple processed materials. Functionality of the resulting algorithm is illustrated by finding optimal designs of a conventional reactor containing vessels with 1 and 5 liter reactants. It is shown that for finding best configurations of the system (suitable for three different reactants) from seven-parameter optimizations only a few dozen/hundred simulations are required.

2. Optimization Technique: RBF Network and Objective Functions

In accordance with [11], the decomposed RBF artificial neural network (ANN) shown in Fig. 1 and denoted as $F: X \rightarrow Y$ works with input vectors $\mathbf{X}_i = [x_1 \ x_2 \ \dots \ x_N]$, where x_1, \dots, x_N are design variables for $i = 1, \dots, P$, and P is the number of input-output pairs of modeling data. The network output vectors are obtained by taking frequency characteristics of an S -parameter over specified frequency range(s) (f_j^1, f_j^2) given by the formula

$$Y_i = \max_{1 \leq k \leq L} \left[\max_{1 \leq j \leq n} \left\{ \left[BW(S(f_j^1 \leq f \leq f_j^2, X_i, \epsilon'_k, \sigma_k), T_j) + 1 \right]^1 \right\} \right], \quad (1)$$

where the function BW (associated with the system's bandwidth) is calculated over the specified frequency interval(s) and is in the range $[0, 1]$, and L is the number of materials for which the system is to be optimized; we maximize over k to use the worst of the L samples with dielectric constant ϵ' and electric conductivity σ . This OF represents a typical practical need of microwave optimization to search not just the minimum of S , but rather the maximum BW in a certain frequency range. The RBF used in the network is a thin plate spline defined as

$$\phi_l^{(i)} = \begin{cases} \|X_i - c_l\|_2^2 \log(\|X_i - c_l\|_2), & \|X_i - c_l\|_2 > 0, \\ 0, & \|X_i - c_l\|_2 = 0, \end{cases} \quad (2)$$

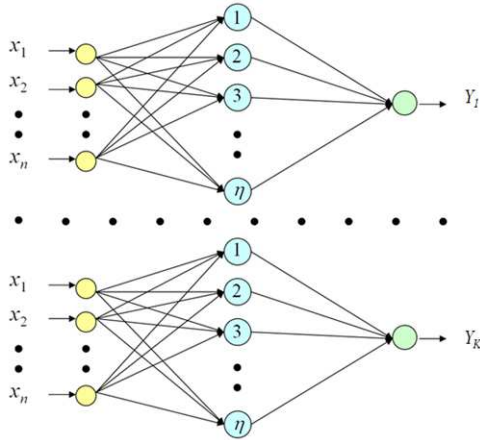


Fig. 1. Architecture of a decomposed RBF ANN with η hidden neuron [11].

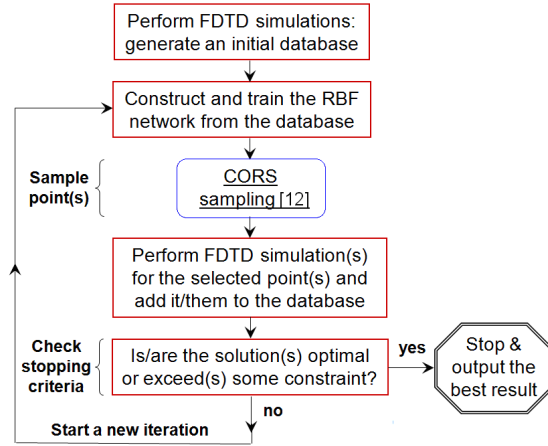


Fig. 2. Flow chart of the RBF optimization algorithm with CORS sampling.

where $l = 1, \dots, N_c$, N_c is the number of RBFs, c_l are the centers of $\phi_l^{(i)}$. The training set is the set of centers chosen. The network is coupled with a linear model, and the weights are constructed by solving the corresponding linear system.

A general description of the algorithm is given in Fig. 2. Given some initial data, we construct and train the RBF network $F(X)$, perform CORS sampling [12], simulate the sampled point, check stopping criteria, and repeat the cycle, if necessary. The most critical part of the algorithm is the choice of the sampled point. CORS sampling balances the goal of finding the minimum with exploring unknown regions of the domain. This is accomplished by selecting a parameter β ($0 \leq \beta \leq 1$) and finding the minimum of $F(X)$, subject to $\|X - X_j\| \geq \beta\Delta$ for $1 \leq j \leq P$, where $\Delta = \max_X(\min_{1 \leq j \leq P} \|X - X_j\|)$.

The algorithm is implemented as a MATLAB code. Data for the network are generated by the 3D conformal FDTD simulator *QuickWave-3D* [13]. All CPU times are given below for a 64-bit Windows XP-based Intel Xeon 3.2-GHz PC.

3. Numerical Results: A MAOS Reactor for Multiple Reactants

Here we consider a microwave system resembling a typical MAOS reactor (Fig. 3). Inside the cylindrical cavity, there is a thin-wall cylindrical vessel with a liquid reactant whose volume V is assumed to be specified. The reactor is excited by a rectangular waveguide which may be offset in the x - and y -directions by f_x and f_y respectively. The vessel also may be offset from the z -axis by v_x and v_y being located a distance s from the top of the cavity. The CAD goal is: given the height of the system H and a set of L reactants to be processed in the reactor, find the configuration of the whole system, i.e., diameter D , internal dimensions of the vessel (d and h), its position in the cavity (s , v_x and v_y), and a position of the waveguide (f_x and f_y), that yields less than $T\%$ of reflected microwave energy (i.e., the reflection coefficient $|S_{11}| < 0.1\sqrt{T}$) in $BW_A = \dots = BW_L$ % of the frequency range (f^1, f^2) for reactants (A), \dots , (L) respectively.

In the first illustration, we solve this problem for the load with $V = 1$ liter and the height of the reactor $H = 300$ mm. We consider $L = 3$ (the materials are specified in Table 1), $T = 1\%$, $BW_A = \dots = BW_C = 100\%$, $f^1 = 2.4$ GHz, and $f^2 = 2.5$ GHz. The seven design variables are allowed to be in the intervals:

$$\begin{aligned} -35 \leq f_x, f_y \leq 35 \text{ mm}, \quad -24 \leq v_x, v_y \leq 24 \text{ mm}, \quad 80 \leq d \leq 110 \text{ mm}, \\ 75 \leq s \leq 110 \text{ mm}, \quad 200 \leq D \leq 300 \text{ mm}. \end{aligned} \tag{3}$$

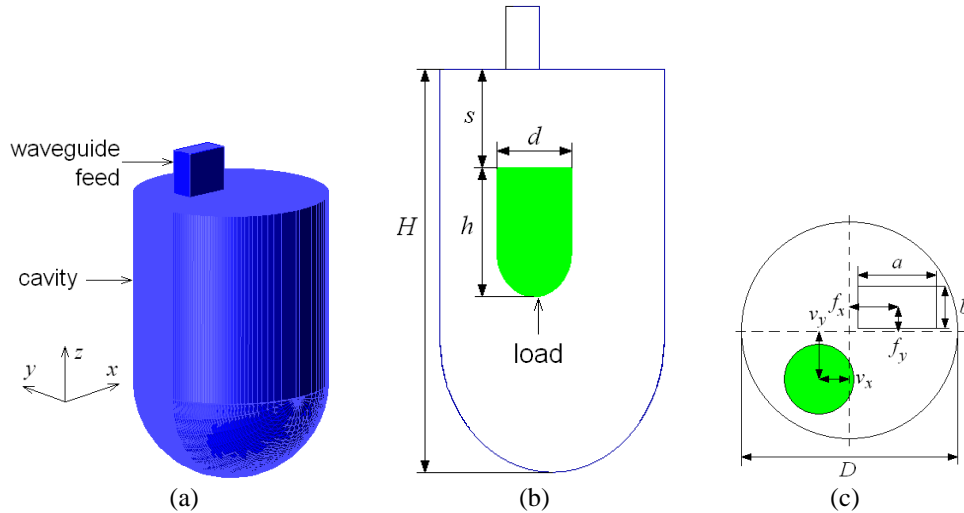


Fig. 3. General 3D view (a) and geometrical parameters of the considered MAOS reactor (b), (c).

Table 1. Material Parameters of the Reactants at 2.45 GHz (Adapted From [1]).

Reactant	Dielectric constant, ϵ'	Electric conductivity, σ (S/m)
(A) Ethyl acetate	6.2	0.1468
(B) Methylene chloride	9.1	0.0582
(C) Ethanol	24.6	0.1808

The procedure starts with generation of an initial database (DB) of 50 random points in the specified domain for each of the three reactants. The underlying FDTD model built with a non-uniform conformal mesh consists of 308,000 to 555,000 cells of maximum 2.7 mm in the reactant to maximum 7 mm in air; the analysis of the system involves 10,000 time-steps (1.7 to 3.0 min of CPU time).

Characteristics of the reflection coefficient in the non-optimized reactor computed for all the reactants yield energy efficiency about 75-85%. Our optimization procedure achieves what seems to be the “best” solution producing 99% efficiency ($BW_A = 79\%$, $BW_B = 83\%$, and $BW_C = 100\%$) with $99 \times 3 = 297$ FDTD analyses (Fig. 4). Corresponding design variables are given in Table 2 and frequency characteristics of $|S_{11}|$ resulting from this geometry are shown in Fig. 5: it is seen that this optimized configuration provides 99% efficiency in the 79% of the 2.4 to 2.5 GHz frequency range for the reactants (A), (B), and (C).

The next example illustrates operation of the proposed optimization technique in situations when scaling up of a successful MAOS process is of key interest. Here we solve the optimization problem for the reactor of the same design and constructed for the same reactants ($L = 3$), but of a 5-liter volume. With $H = 513$ mm, $T = 1\%$, $BW_A = \dots = BW_C = 90\%$, $f^1 = 2.4$ GHz, and $f^2 = 2.5$ GHz, the seven design variables are allowed to be in the intervals:

$$\begin{aligned} -85 \leq f_x, f_y \leq 85 \text{ mm}, \quad -50 \leq v_x, v_y \leq 50 \text{ mm}, \quad 159 \leq d \leq 172 \text{ mm}, \\ 80 \leq s \leq 300 \text{ mm}, \quad 340 \leq D \leq 510 \text{ mm} \end{aligned} \quad (4)$$

Similarly, the procedure starts with an initial DB of 50 random points in the domain (4). The model consists of 1,380,000 to 2,394,000 cells of maximum 3 mm in the reactant to maximum 7 mm in air – due to a larger reactant’s volume the domain discretized with 3 mm cells is also larger. Likewise, the analysis requires 10,000 time-steps (8.6 to 14.4 min of CPU time).

In the case of a 5 liter reactant, as seen from Fig. 6, the best solution ($BW_A = 80\%$, $BW_B = 77\%$, and $BW_C = 70\%$) is achieved with 119 DB points, but for the preceding very close result

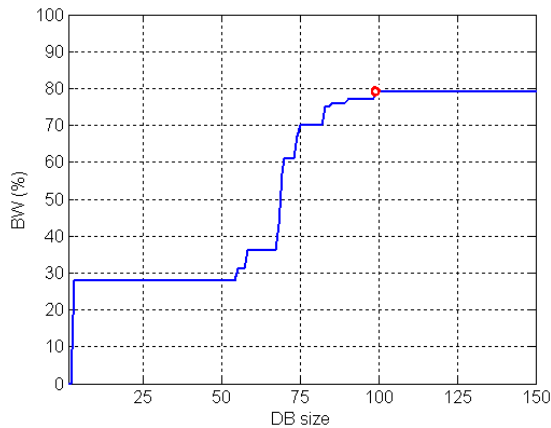


Fig. 4. Optimization convergence for the MAOS reactor designed for three 1-liter reactants (A), (B), (C); (○) marks the 99-point solution.

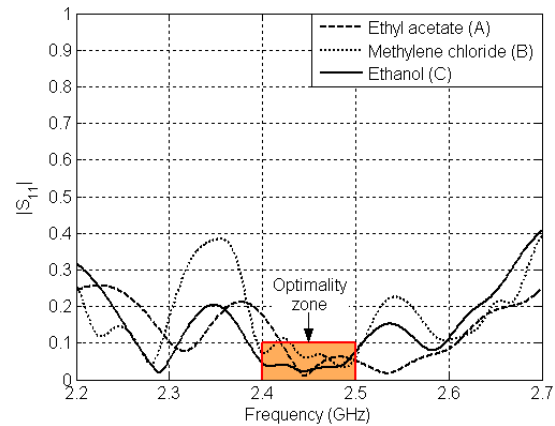


Fig. 5. Reflection coefficient in the MAOS reactor optimized for three 1-liter reactants (A), (B), (C).

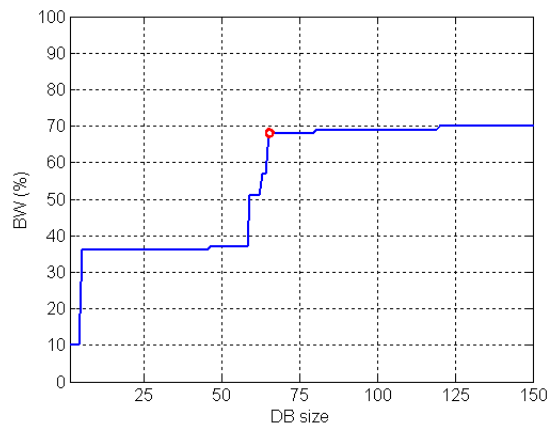


Fig. 6. Optimization convergence for the MAOS reactor designed for three 5-liter reactants (A), (B), (C); (○) marks the 68-point solution.

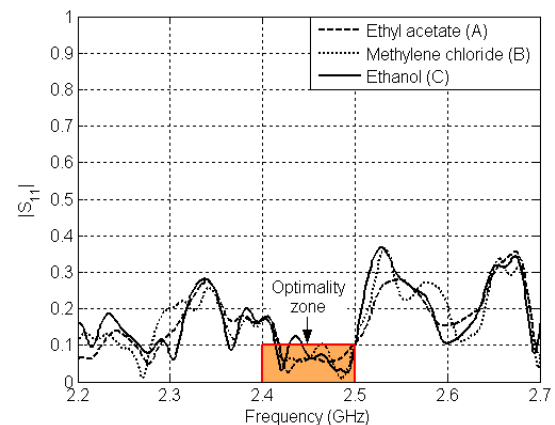


Fig. 7. Reflection coefficient in the MAOS reactor optimized for three 5-liter reactants (A), (B), (C).

Table 2. A MAOS Reactor (Fig. 3): Configurations Optimized for Reactants (A), (B), and (C).

Reactant's volume	f_x , mm	f_y , mm	v_x , mm	v_y , mm	d , mm	s , mm	D , mm	No of DB points/No of FDTD analyses
1 liter [(A)/(B)/(C)]	-23.4	11.0	-5.3	20.0	108.0	94.8	209.0	99 / 297
5 liter [(A)/(B)/(C)]	75.8	83.2	50.0	-36.0	164.0	145.4	395.2	65 / 195

(with the worst $BW_C = 68\%$) the procedure needs only 65 points (195 analyses). Corresponding design variables are given in Table 2 and $|S_{11}|$ characteristics resulting from this geometry are shown in Fig. 7: this optimized configuration provides 99% energy efficiency for all three reactants in the 70% of the 2.4 to 2.5 GHz frequency range.

It is seen that the optimized geometry of the 5 liter reactor is fairly different from a proportionally increased one of the 1 liter system, and the “qualities” of both best solutions are not alike. This appears to be consistent with theoretically expected (and observed in many experiments, see, e.g., [10]) strongly non-linear alterations in reactor's performance with variations of its geometrical parameters and materials characteristics of the reactants.

4. Conclusions

An original modeling-based approach for CAD and scaling up of MAOS reactors has been proposed. The technique allows for getting the geometry of a large-scale reactor that guarantees highest possible energy efficiency for different reactants. The technique of RBF ANN optimization backed by full-wave (3D conformal FDTD) simulations has been applied to a conventional microwave reactor to find its best design for processing of 1- and 5-liter reactants. It has been shown that, with the developed technique, seven-parameter optimization of energy efficiency of this system takes only 297 and 195 FDTD analyses (or about 16 and 37.5 h of CPU time), respectively. The proposed technique is straightforwardly applicable to optimization of the systems with reactants whose material parameters strongly depend on temperature.

The results presented in this paper stay in strong favor of CAD of high-productive large-scale MAOS reactors and suggest that application of the developed modeling-based optimization procedure in efficient designing of systems of microwave chemistry may be useful and viable.

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