

Power Changes of Electromagnetic Waves Across the Temporal Boundary in Simple Polar-Molecule Reactions

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Abstract – Microwave-assisted chemical reactions have been widely used, but the inhomogeneous heating limits further applications. The aim of this paper is to investigate the power transfer behavior in the simple polar-molecule reactions whose polarization changes with the proceeding of the reactions. At the temporal boundary, based on the continuity of charge and flux and the equivalent transmission line approach of the simple polar-molecule reactions, we discover the power changes in the reactions. The numerical results are in agreement with the theory of the temporal boundary. When the time scale of the component concentration variation is smaller than the wave period, the polarization is not continuous at the temporal boundary. The impedance of the reactions across the temporal boundary changes, and the reflection occurs. Moreover, when the dielectric property of the reactions decreases, the power of the waves increases after the temporal boundary and the waves experience a net energy gain. The results may be helpful in disclosing the non-uniform electromagnetic energy distribution in chemical reactions.

Index Terms – Electromagnetic power propagation, power changes, a temporal boundary, simple polar-molecule reactions.

I. INTRODUCTION

Microwaves have been used increasingly in chemical engineering for a series of advantages, such as improvement in reaction rates, yields, and energy saving [1–3]. However, inhomogeneous heating and hot spots have been limiting the utilization of microwaves in chemical engineering [4, 5]. In order to study these problems, it is essential to investigate the behavior of electromagnetic power transfer in chemical reactions.

The electromagnetic energy propagation through general media with dispersion and loss has been

studied extensively for decades [6, 7]. The previous studies are mainly focused on electromagnetic media characterized by permittivity and permeability. However, most of chemical reactions are non-stationary media, of which dielectric properties are different from the media reported before [8–10]. Recently, the time-varying and dispersive characteristics of chemical reactions have been demonstrated theoretically [11]. Perhaps, the most outstanding features of chemical reactions are time-varying characteristics, which cause changes in the amplitude and frequency of electromagnetic waves. And the reflection and transmission of electromagnetic waves at a temporal boundary have been discussed in chemical reactions [12]. What is more important, the internal reflections caused by time-varying characteristics lead to the non-uniform distribution of electromagnetic waves in chemical reactions. To the best of our knowledge, the effects of time-varying characteristics on electromagnetic energy in chemical reactions are barely mentioned. It is the first time that the simple polar-molecule reactions are chosen as the research object to study the effect of the time-varying characteristics on the power changes. Moreover, the power changes at such temporal characteristics will further stimulate an intense investigation in the non-uniform distribution of electromagnetic energy in spatially homogenous chemical reactions. In order to obtain the power changes across the temporal boundary in the homogenous reactions, the impact of component concentration variation on the conditions and trends of power changes are shown for the first time.

In recent years, the transmission line theory has evolved into a very powerful modeling tool to analyze energy flux in dynamic systems, such as the mechanical and hydraulic systems [13–15]. The unit equivalent circuit of a transmission line can be connected to the resistors, inductors, and capacitors under constraints of a

physical meaning of systems to be modeled [16, 17]. In this approach, electromagnetic field quantities are substituted by the analogous incident and reflected voltage and current propagating through this unit circuit [18, 19]. This conceptually new approach greatly simplifies calculations by the definite physical meaning of circuit elements in the unit circuit. And the fundamental electromagnetic properties of the reactions are reviewed by a unit equivalent circuit approach [20]. The equivalent transmission line approach provides an efficient analysis tool for the power transfer in chemical reactions.

In this letter, we investigate the unique properties of the power transfer through homogenous polar-molecule reactions based on an equivalent transmission line approach. Then the propagation of electromagnetic energy in the reactions is simulated to validate the theoretical results.

II. POWER CHANGES ACROSS A TEMPORAL BOUNDARY IN REACTIONS

The power of electromagnetic waves in media can often be calculated by permittivity. However, many chemical reactions are non-equilibrium process. During this process, the concentrations of reactants and products are changing with time. Based on the modified Smoluchowski–Debye equation, the polarization expression of polar-molecule reactions is indicated by the response function in the time domain. The expression of the polarization in polar-molecule reactions is given as

$$\mathbf{P}(t) = \sum_i \varphi_{A_i}(t) * [c_{A_i}(t)\mathbf{E}(t)] + \sum_j \varphi_{B_j}(t) * [c_{B_j}(t)\mathbf{E}(t)].$$

Here, * denotes the convolution. $\varphi_{A_i}(t)$ and $\varphi_{B_j}(t)$ represent the rotational diffusion properties of the reactant and product, while $c_{A_i}(t)$ and $c_{B_j}(t)$ denote the concentrations of the reactant and product, respectively [10]. To make the polarization expression easily, the simple polar-molecule reactions are considered, in which only one component (X_i) has the polarization

$$\mathbf{P}_{X_i}(t) = \varphi_{X_i}(t) * [c_{X_i}(t)\mathbf{E}(t)]. \quad (1)$$

$\varphi_{X_i}(t)$ and $c_{X_i}(t)$ are the rotational diffusion function and concentration of X_i . * represents the convolution. $\varphi_{X_i}(t) = \epsilon_0 \chi_{X_i} \exp(-t/\tau_{X_i})/\tau_{X_i}$, where χ_{X_i} is the electric susceptibility, ϵ_0 is the vacuum permittivity, and τ_{X_i} is the dielectric relaxation time. X_i denotes the reactants or products [10]. Furthermore, the reactions are considered to be more general media of composite dispersive and time-varying characteristics [11]. To show the effects of time-varying characteristics on the power transfer, we consider an instant t_0 as the temporal boundary in chemical process. Figure 1(a) shows the polarization of reaction changes from $\mathbf{P}_{X_i}(t_0^-)$ to $\mathbf{P}_{X_i}(t_0^+)$ between a temporal boundary t_0 , which lead to the intrinsic impedance of the reactions varied from $\eta(t_0^-)$ to $\eta(t_0^+)$.

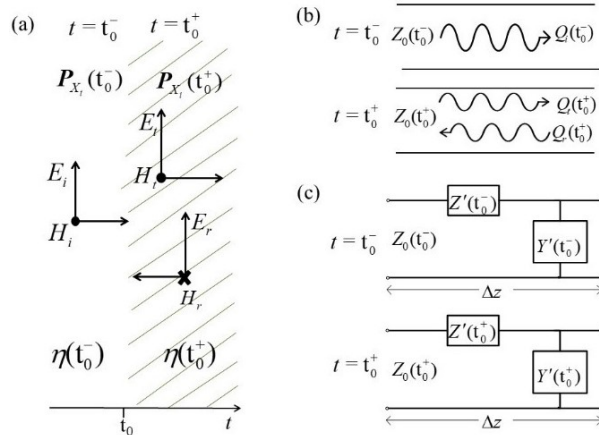


Fig. 1. (a) Propagation processes at the temporal boundary in space-invariant reactions. (b) Schematic of time-varying transmission line. (c) The equivalent circuit unit.

To indicate the features of power changes resulting from the time-varying characteristics of the reactions, the reactions are treated as spatially homogeneous and isotropic nonmagnetic media, being without free charges and currents. The transmission line theory can be employed to explain the electromagnetic propagation behavior in media [21, 22]. The homogenous reactions can be represented by an equivalent transmission line (continuous and invariant along the direction of propagation) to gain an immediate insight into the fundamental properties of reactions. The time-varying transmission line represented in Figure 1(b) corresponds to the model of Figure 1(a). We denote the charge and flux before t_0 with $Q_i(t_0^-)$ and $\phi_i(t_0^-)$. After t_0 , the transmitted and reflected charges are denoted by $Q_t(t_0^+)$ and $Q_r(t_0^+)$, respectively, with a similar notation for the fluxes.

Figure 1(c) shows the unit circuit of the transmission line presenting a per-unit length for media as described by eqn (1). In such a transmission line circuit, the magnetization and polarization are represented by the distributed series impedance (Z') and shunt admittance (Y') per unit length, respectively [16, 17], according to the constitutive relation of the reactions. Since the time-varying characteristics of the reactions, the principle of the time-varying transmission line is to make its characteristic parameters, such as impedance and admittance, being time changing. $Z'(t_0^-)$ and $Z'(t_0^+)$ denote the impedances before and after the temporal boundary located at t_0 , similarly for the admittances $Y'(t_0^-)$ and $Y'(t_0^+)$. The transmission line's characteristic impedance ($Z_0 = \sqrt{Z'/Y'}$) changes from $Z_0(t_0^-)$ to $Z_0(t_0^+)$ at $t = t_0$, which correspond to the reactions' intrinsic impedance. In its unit equivalent circuit, the series

impedance (Z') and shunt admittance (Y') can be represented as the combination of equivalent time-varying resistance ($R(t)$), conductance ($G(t)$), capacitance ($C(t)$), and inductance ($L(t)$) of per-unit length, which can be derived mathematically by Maxwell equations and the constitutive relations of the reactions. The equivalent capacitance changes from $C(t_0^-)$ to $C(t_0^+)$ at $t = t_0$, with a similar notation for the inductance. For a time-varying equivalent circuit, the relationships between charge and voltage is $Q(z, t) = C(t)u(z, t)$ as well as the flux and current is $\phi(z, t) = L(t)i(z, t)$ [23, 24]. Thus, the instantaneous power is given by

$$P(t) = u(t)i(t) = \frac{u^2(t)}{Z_0(t)} = \frac{Q^2(t)}{C^2(t)Z_0(t)}. \quad (2)$$

$\nabla \times \mathbf{E} = -\partial \mathbf{B} / \partial t$ and $\nabla \times \mathbf{H} = \partial \mathbf{D} / \partial t$ indicate that the electric displacement (\mathbf{D}) and magnetic induction (\mathbf{B}) require the continuity at a temporal boundary [25, 26]. As the charge $Q = \int \mathbf{D} \cdot d\mathbf{S}$ and the flux $\phi = \int \mathbf{B} \cdot d\mathbf{S}$, the charge (Q) and flux (ϕ) are conserved at a temporal boundary t_0 [27]. Accordingly, we have

$$Q_i(t_0^-) = Q_r(t_0^+) + Q_t(t_0^+) \quad (3a)$$

and

$$\phi_i(t_0^-) = -\phi_r(t_0^+) + \phi_t(t_0^+). \quad (3b)$$

Thus, the ratio of the reflected power (P_r) and transmitted power (P_t) to the incident power (P_i) at t_0 are derived as

$$\frac{P_r}{P_i} = \frac{C^2(t_0^-)Z_0(t_0^-)}{C^2(t_0^+)Z_0(t_0^+)} \frac{1}{4} \left(1 - \frac{C(t_0^+)Z_0(t_0^+)}{C(t_0^-)Z_0(t_0^-)} \right)^2 \quad (4a)$$

and

$$\frac{P_t}{P_i} = \frac{C^2(t_0^-)Z_0(t_0^-)}{C^2(t_0^+)Z_0(t_0^+)} \frac{1}{4} \left(1 + \frac{C(t_0^+)Z_0(t_0^+)}{C(t_0^-)Z_0(t_0^-)} \right)^2. \quad (4b)$$

To get a deeper insight, we add the eqn (4a) and (4b) and obtain

$$\frac{P_r + P_t}{P_i} = \frac{C(t_0^-)}{C(t_0^+)} \frac{1}{2} \left(\frac{C(t_0^-)Z_0(t_0^-)}{C(t_0^+)Z_0(t_0^+)} + \frac{C(t_0^+)Z_0(t_0^+)}{C(t_0^-)Z_0(t_0^-)} \right). \quad (5)$$

Eqn (4a) and (4b) indicate that the reflected and transmitted powers change due to an abrupt change of impedance. When the time scale of the component concentration variation is greater than the wave period, the time-varying characteristics of the reactions can be neglected and $\mathbf{P}_{X_i}(t_0^-) = \mathbf{P}_{X_i}(t_0^+)$ [11]. The unit parameters of the transmission line circuit do not change at t_0 , that is, $C(t_0^-) = C(t_0^+)$ and $Z_0(t_0^-) = Z_0(t_0^+)$. As shown in eqn (4a) and (4b), $P_r = 0$ and $P_t = P_i$ are obtained subsequently. Thus, $(P_r + P_t)/P_i = 1$ by eqn (5), indicating conservation of energy. It is important to realize that in the absence of the time-varying property of the reactions, $\mathbf{P}_{X_i}(t_0^-) = \mathbf{P}_{X_i}(t_0^+)$ and the temporal boundary is switched off. The reflectivity vanishes and the energy of the waves is conserved.

On the other hand, while the time scale of the component concentration variation is smaller than the

wave period, the time-varying characteristics of the reactions are obvious, and $\mathbf{P}_{X_i}(t_0^-) \neq \mathbf{P}_{X_i}(t_0^+)$ [11]. Thus, $C(t_0^-) \neq C(t_0^+)$ and $Z_0(t_0^-) \neq Z_0(t_0^+)$. The incident power is changed into two parts: the reflected power (P_r) and the transmitted power (P_t). $(P_r + P_t)/P_i \neq 1$ by eqn (5), indicating the non-conservation of energy for the waves. As the energy is conserved for the whole waves and media, eqn (5) can be interpreted as an energy exchange between the waves and the time-varying media [28–30]. Furthermore, an energy exchange occurs between the waves and reactions, which has been demonstrated to lead to either amplification or attenuation of electromagnetic waves depending on whether the dielectric property of the reactions decrease or increases with time [11, 31]. More specially, if the relaxation time of the reactions is much smaller, the characteristic impedance $Z_0(t_0^-)$ and $Z_0(t_0^+)$ are assumed as real numbers approximately. Thus, $\frac{1}{2} \left(\frac{C(t_0^-)Z_0(t_0^-)}{C(t_0^+)Z_0(t_0^+)} + \frac{C(t_0^+)Z_0(t_0^+)}{C(t_0^-)Z_0(t_0^-)} \right) \geq 1$. Particularly, when $C(t_0^-) > C(t_0^+)$, we can get $P_r + P_t > P_i$. Here, it is worth remarking that the sum of the reflected and transmitted power is greater than the incident power. Thus, the reactions bearing the time-varying characteristic act as a source of energy for electromagnetic waves. This fact should be considered in microwave heating to achieve the homogeneous power transfer in the reactions.

To clarify the physical meaning of the derived expression, parametric phenomena of electromagnetic waves in active media can be realized by a time modulation of media parameters [32]. In fact, the number of dipoles would be changed with the proceeding of chemical reactions. It can be inferred that the equivalent time-varying capacitance of the reaction is weakened with the decrease of dipoles' quantity; that is, $C(t_0^-) > C(t_0^+)$. What is more, the electromagnetic waves propagate in the reactions bearing time-varying characteristics, just as if they were in contact with a reservoir (time-dependent background media) [33, 34]. The decrease of the time-varying capacitor leads to reductions of the reservoir energy. This essentially leads to an energy flow from the reservoir to the electromagnetic waves. Therefore, the time-varying capacitors through the bounded discontinuities have an additional contribution as active circuit elements to the electromagnetic power.

III. RESULTS AND DISCUSSIONS

As discussed above, to indicate the unique features of power propagation originating from the time-varying characteristics of the reactions, the effects of component concentration variation on the transmitted power are discussed by the numerical simulations. We consider a monochromatic wave incident into the reactions, so as to understand the influence of the time-varying

characteristics on the transmitted and reflected powers. For the electric field, the single cycle sine wave $E_i(t)$ is employed as

$$E_i(t) = \begin{cases} \sin(2\pi \times 5 \times 10^9 \times t) & t \leq 0.1\text{ns} \\ 0 & t > 0.1\text{ns} \end{cases} \quad (6)$$

In the calculation, the reactions with the electric susceptibility $\chi_{X_i}=10$ and the dielectric relaxation time $\tau_{X_i}=1\text{fs}$ are used. This paper selects an instant t_0 in chemical process and the temporal boundary t_0 is set as 2 ns. To investigate the power changes in the simple polar-molecule reactions, without loss of generality, the reactions are considered as the first-order reaction.

A. Power changes when X_i is the reactant

The normalized concentration $c_{X_i}(t) = \exp(-K_{rate}t)$ can be obtained when X_i is the reactant. K_{rate} is the rate constant of the reactions. The electromagnetic powers in the simple polar-molecule reactions are shown in Figure 2.

When K_{rate} is chosen as 0s^{-1} , $1 \times 10^3\text{s}^{-1}$, and $1 \times 10^8\text{s}^{-1}$, power changes cannot be seen in Figure 2(a)–(c). We find that the reflected power (P_r) is zero and the transmitted power (P_t) is matched with the incident power (P_i). As the time scale of the concentration variation is greater than the wave period, the polarization $P_{X_i}(t)$ is continuous at t_0 . Since the impedance of the reactions does not change at t_0 , no reflection occurs. Moreover, using this condition employed in the unit equivalent circuit of the reactions, $P_r = 0$ and $P_t = P_i$ are obtained by eqn (4a) and (4b), indicating that there are no power changes. When $K_{rate} = 1 \times 10^{11}\text{s}^{-1}$, the reflected power (P_r) is observed clearly, and the transmitted power (P_t) is not equal to the incident power (P_i) in Figure 2(d). As the time scale of the concentration variation is smaller than the wave period, i.e., $c_{X_i}(t)$ changes rapidly with time, the polarization $P_{X_i}(t)$ is not continuous at the temporal boundary, inducing the discontinuous impedance change. For this situation, $P_r \neq 0$ and $P_t \neq P_i$ are obtained by eqn (4a) and (4b), and $(P_r + P_t)/P_i \neq 1$ by eqn (5) indicates power changes. Another phenomenon of interest is that the transmitted power is greater than the incident power.

B. Power changes when X_i is the product

On the other hand, when X_i is the product, the expression of $c_{X_i}(t) = 1 - \exp(-K_{rate}t)$ can be achieved and the electromagnetic powers in the simple polar-molecule reactions are shown in Figure 3. The reflected power is not observed as $K_{rate} = 0\text{s}^{-1}$, $1 \times 10^3\text{s}^{-1}$, and $1 \times 10^8\text{s}^{-1}$ in Figure 3(a)–(c). And we cannot see the power changes. When $K_{rate} = 1 \times 10^{11}\text{s}^{-1}$, the reflected power can be observed, and the transmitted power (P_t) is smaller than the incident power (P_i) in Figure 3(d). We can find the power changes across the temporal boundary in the reactions in Figure 3(d).

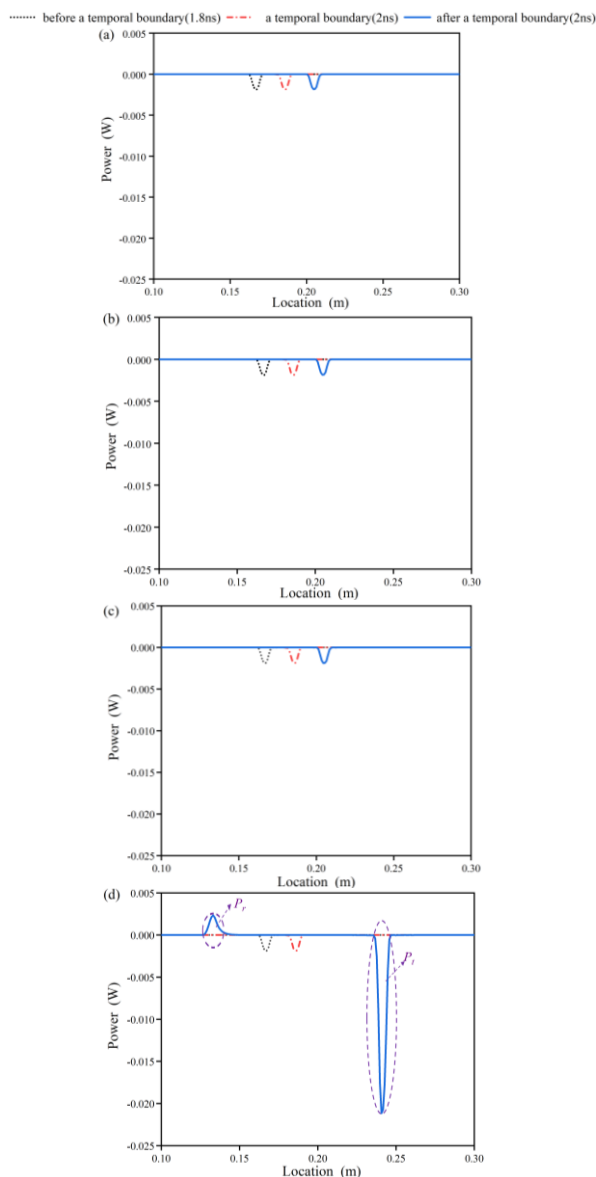


Fig. 2. The electromagnetic powers in the simple polar-molecule reactions when X_i is the reactant. (a) $K_{rate} = 0\text{s}^{-1}$. (b) $K_{rate} = 1 \times 10^3\text{s}^{-1}$. (c) $K_{rate} = 1 \times 10^8\text{s}^{-1}$. (d) $K_{rate} = 1 \times 10^{11}\text{s}^{-1}$.

A temporal variation of the dielectric property is created because the component concentrations change in chemical process. When X_i is the reactant, the component concentration of X_i decreases gradually in the reactions. It can be inferred that the dielectric property is weakened with the decrease of dipoles' quantity when X_i is the reactant. For this situation, the equivalent time-varying capacitance of the reaction decreases as the dielectric property becomes smaller. Based on eqn (5), $P_t + P_r > P_i$ is obtained, indicating the increase of power across the temporal boundary t_0 in the reactions.

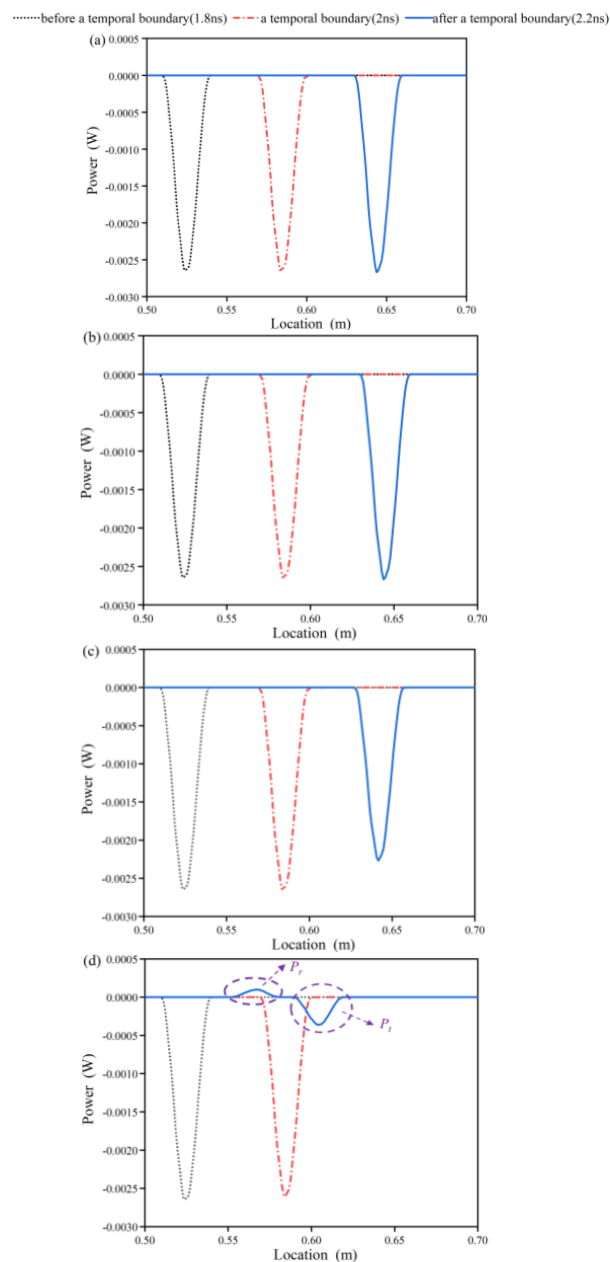


Fig. 3. The electromagnetic powers in the simple polar-molecule reactions when X_i is the product. (a) $K_{rate} = 0s^{-1}$. (b) $K_{rate} = 1 \times 10^3s^{-1}$. (c) $K_{rate} = 1 \times 10^8s^{-1}$. (d) $K_{rate} = 1 \times 10^{11}s^{-1}$.

When the reactions decrease media's dielectric property, the power of electromagnetic waves increases after the transition, and, thus, the electromagnetic waves experience a net energy gain [35]. This should come as a surprise since decreasing the dielectric property of the reactions is an energy-releasing process. For the reverse case, when X_i is the product, the opposite occurs and the wave experiences a net energy loss because increas-

ing the dielectric property of the reactions consumes energy.

IV. CONCLUSION

Based on the continuity of charge and flux at a temporal boundary and the equivalent transmission line approach of the simple polar-molecule reactions, this paper focuses on the power changes across the temporal boundary. According to the time-varying characteristics of the reactions, the effects of component concentration variation on the power changes are discussed. When the time scale of the concentration variation is smaller than the wave period, the polarization of the reactions is not continuous at the temporal boundary, inducing the discontinuous impedance change. Importantly, when the reactions decrease media's dielectric property, the transmitted power is greater than the incident power; thus, the wave experiences a net energy gain. These results may be helpful in disclosing the non-uniform distribution of electromagnetic energy in chemical reactions.

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