FRACTAL DIFFUSION-REACTION MODEL FOR A POROUS ELECTRODE

by

Ling LIN[a,b] and Yun QIAO[a,b]*

[a] Zhejiang Fashion Institute of Technology, Ningbo, China
[b] School of Economics and Management, Huanghe Jiaotong University, Wuzhi Town, Jiaozuo City, Henan Province, China

Original scientific paper
https://doi.org/10.2298/TSCI191212026L

Fractal modifications of Fick's laws are discussed by taking into account the electrode's porous structure, and a fractal derivative model for diffusion-reaction process in a thin film of an amperometric enzymatic reaction is established. Particular attention is paid to giving an intuitive grasp for its fractal variational principle and its solution procedure. Extremely fast or extremely slow diffusion process can be achieved by suitable control of the electrode's surface morphology, a sponge-like surface leads to an extremely fast diffusion, while a lotus-leaf-like uneven surface predicts an extremely slow process. This paper sheds a bright light on an optimal design of an electrode's surface morphology.

Key words: *approximate solution, geometric potential theory, fractal calculus, amperometric enzymatic reaction, fractal variational principle*

Introduction

A non-linear problem arising in an amperometric enzymatic reaction is generally modelled by a differential equation [1-6], its variational model was rarely studied though it has obvious advantages over its differential partner. Due to the difficulty in the establishment of a needed variational formulation, the variational approach has not yet widely adopted in chemistry community, now the condition is completely changed, we can use the semi-inverse method [7-13] to find easily a variational principle for the studied problem.

Additionally the differential model cannot describe the effect of electrode's porous structure on the diffusion-reaction process, and this paper is to establish a fractal derivative model to study the process using a fractal variational principle.

Diffusion-reaction equation

Fick's laws are widely used to describe various diffusion process [14], the First law implies that the diffusion flux can be expressed:

$$\mathbf{J} = D\nabla s \tag{1}$$

where s is the concentration, D – the diffusion coefficient, $\mathbf{J}$ – the flux. The Second law:

$$\frac{\partial s}{\partial t} = \nabla \mathbf{J} \tag{2}$$

* Corresponding author, e-mail: xinranyun@163.com

So we have the following diffusion equation:

$$\frac{\partial s}{\partial t} = \nabla(D\nabla \mathrm{s}) \tag{3}$$

For 1-D diffusion process, we have:

$$\frac{\partial s}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial s}{\partial x}\right) \tag{4}$$

The reaction-diffusion equation [14-17]:

$$\frac{\partial s}{\partial t} = \nabla(D\nabla \mathrm{s}) - v(t,x,y,z) \tag{5}$$

where v is the reaction velocity, which can be described by the Michaelis-Menton equation:

$$v(t,x,y,z) = \frac{Ms}{1+\dfrac{s}{K_M}} \tag{6}$$

where K and K_M are constants. The 1-D diffusion-reaction equation:

$$\frac{\partial s}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial s}{\partial x}\right) - \frac{Ms}{1+\dfrac{s}{K_M}} \tag{7}$$

Fractal Fick's law

The Fick's laws cannot describe the effect of porous structure on the diffusion process. In Fick's laws, the concentration is a function of time and co-ordinatess $s(t, x, y, z)$, however, the diffusion might be anomalous, it is not differentiable with respect to time and space, but it is differentiable with respect to t^δ, x^α, y^β, z^γ, where δ, α, β, and γ can be explained as fractal dimensions which will be discussed later. That means the concentration is a function $s(t^\delta, x^\alpha, y^\beta, z^\gamma)$ instead of $s(t, x, y, z)$.

Experiment also reveals that the concentration change across Δt or Δx or Δy or Δz can be expressed [18-23]:

$$\Delta s \propto (\Delta t)^\delta \tag{8}$$

$$\Delta s \propto (\Delta x)^\alpha \tag{9}$$

$$\Delta s \propto (\Delta y)^\beta \tag{10}$$

$$\Delta s \propto (\Delta \mathrm{z})^\gamma \tag{11}$$

where Δt, Δx, Δy, and Δz are the smallest scales in space and time, respectively, any phenomena measured on a scale smaller than Δt, Δx, Δy, or Δz are ignored. In fractal calculus, we always assume that $\Delta x \neq 0$, $\Delta y \neq 0$, $\Delta z \neq 0$, and $\Delta t \neq 0$, so Δx, Δy, and Δz can be explained as the smallest porous sizes in x-, y- and z-directions, respectively, Δt can be explained as the smallest time scale for the concentration change across Δx, Δy, or Δz, and δ, α, β, and γ can be understood as the fractal dimensions in time and space, respectively.

To understand the scaling laws given in eqs. (8)-(11), we consider an animal working along a coastline with a stride of Δx. Any discontinuity within Δx is completely ignored. If there is a river along the coastline, and its width is smaller than Δx, then the river can be

ignored. However, if Δx is smaller than the river width, the animal has to go along the river to the opposite of the river, so the average velocity of the animal follows an allometric scaling law $\bar{u} \propto (\Delta x)^{\alpha}$, where α is the fractal dimension of the coastline.

According to the aforementioned analysis, Fick's First law and second law can be modified:

$$\mathbf{J} = D\nabla^{(\alpha,\beta,\gamma)}s \tag{12}$$

$$\frac{\partial s}{\partial t^{\delta}} = \nabla^{(\alpha,\beta,\gamma)}[D\nabla^{(\alpha,\beta,\gamma)}s] \tag{13}$$

where the gradient operator in a fractal space is defined:

$$\nabla^{(\alpha,\beta,\gamma)}s = \frac{\partial s}{\partial x^{\alpha}}\mathbf{i} + \frac{\partial s}{\partial y^{\beta}}\mathbf{j} + \frac{\partial s}{\partial z^{\gamma}}\mathbf{k} \tag{14}$$

where $\partial s/\partial x^{\alpha}$, $\partial s/\partial y^{\beta}$, $\partial s/\partial z^{\gamma}$, and $\partial s/\partial t^{\delta}$ are fractal derivatives defined:

$$\frac{\partial s}{\partial x^{\alpha}}(t^{\delta}, x_0^{\alpha}, y^{\beta}, z^{\delta}) = \Gamma(1+\alpha)\lim_{\substack{x-x_0\to\Delta x\\ \Delta x\neq 0}} \frac{s(t^{\delta}, x^{\alpha}, y^{\beta}, z^{\delta}) - s(t^{\delta}, x_0^{\alpha}, y^{\beta}, z^{\delta})}{(x-x_0)^{\alpha}} \tag{15}$$

$$\frac{\partial s}{\partial y^{\beta}}(t^{\delta}, x^{\alpha}, y_0^{\beta}, z^{\delta}) = \Gamma(1+\beta)\lim_{\substack{y-y_0\to\Delta y\\ \Delta y\neq 0}} \frac{s(t^{\delta}, x^{\alpha}, y^{\beta}, z^{\delta}) - s(t^{\delta}, x^{\alpha}, y_0^{\beta}, z^{\delta})}{(y-y_0)^{\beta}} \tag{16}$$

$$\frac{\partial s}{\partial z^{\gamma}}(t^{\delta}, x^{\alpha}, y^{\beta}, z_0^{\delta}) = \Gamma(1+\gamma)\lim_{\substack{z-z_0\to\Delta z\\ \Delta z\neq 0}} \frac{s(t^{\delta}, x^{\alpha}, y^{\beta}, z^{\delta}) - s(t^{\delta}, x^{\alpha}, y^{\beta}, z_0^{\delta})}{(z-z_0)^{\gamma}} \tag{17}$$

$$\frac{\partial s}{\partial t^{\delta}}(t_0^{\delta}, x^{\alpha}, y^{\beta}, z^{\delta}) = \Gamma(1+\delta)\lim_{\substack{z-z_0\to\Delta z\\ \Delta z\neq 0}} \frac{s(t^{\delta}, x^{\alpha}, y^{\beta}, z^{\delta}) - s(t_0^{\delta}, x^{\alpha}, y^{\beta}, z^{\delta})}{(t-t_0)^{\delta}} \tag{18}$$

The diffusion-reaction equation becomes:

$$\frac{\partial s}{\partial t^{\delta}} = \nabla^{(\alpha,\beta,\gamma)}[D\nabla^{(\alpha,\beta,\gamma)}s] - v(t^{\delta}, x^{\alpha}, y^{\beta}, z^{\gamma}) \tag{19}$$

Michaelis-Menton equation should be modified:

$$v(t^{\delta}, x^{\alpha}, y^{\beta}, z^{\gamma}) = \frac{Ms^n}{1+\dfrac{s^n}{K_M}} \tag{20}$$

where n is a constant.

Its 1-D model:

$$\frac{\partial s}{\partial t^{\delta}} = \frac{\partial}{\partial x^{\alpha}}\left(D\frac{\partial s}{\partial x^{\alpha}}\right) - \frac{Ms^n}{1+\dfrac{s^n}{K_M}} \tag{21}$$

The steady diffusion-reaction equation:

$$\frac{\partial}{\partial x^{\alpha}}\left(D\frac{\partial s}{\partial x^{\alpha}}\right) - \frac{Ms^n}{1+\dfrac{s^n}{K_M}} = 0 \tag{22}$$

Fractal variational principle

In this paper we study the steady diffusion-reaction process happened in a thin film of an amperometric enzyme electrode [1-3]. The governing equation:

$$\frac{\partial}{\partial x^{\alpha}}\left(\frac{\partial s}{\partial x^{\alpha}}\right)-\frac{ks^{n}}{1+\sigma s^{n}}=0 \tag{23}$$

with the boundary conditions

$$s'(0)=0,\ \ s(L^{\alpha})=ks_{\infty} \tag{24}$$

where $k = M/D$, $\sigma = 1/K_M$, L – the film thickness, ks_∞ – the substrate's surface concentration in the film, and α – the fractal dimension describing the porous structure of the electrode. A nanoscale porosity on the surface has a high geometric potential [24-27], which can greatly affect the diffusion-reaction process. The value of α plays an important role in the process, and can be used to optimize the process.

In order to elucidate the solution properties of the fractal diffusion-reaction process, we want to establish a fractal variational formulation [12,13, 28, 29]. In order to make the paper concise, we consider a traditional case for $n = 1$, the fractal variational formulation for eq. (23) can be established by the semi-inverse method [7-13]:

$$J(s)=\int_{0}^{L^{\alpha}}\left\{\frac{1}{2}\left(\frac{\mathrm{d}s}{\mathrm{d}x^{\alpha}}\right)^{2}+\frac{k}{\sigma}\left[s-\frac{1}{\sigma}\ln(1+\sigma s)\right]\right\}\mathrm{d}x^{\alpha} \tag{25}$$

Proof. In the fractal variational principle given in eq. (25), the fractal Lagrange function:

$$H=\frac{1}{2}\left(\frac{\mathrm{d}s}{\mathrm{d}x^{\alpha}}\right)^{2}+\frac{k}{\sigma}\left[s-\frac{1}{\sigma}\ln(1+\sigma s)\right] \tag{26}$$

The fractal Euler-Lagrange equation of eq. (25):

$$\frac{\partial H}{\partial s}-\frac{\mathrm{d}}{\mathrm{d}x^{\alpha}}\left(\frac{\partial H}{\partial\dfrac{\mathrm{d}s}{\mathrm{d}x^{\alpha}}}\right)=0 \tag{27}$$

or

$$-\frac{\mathrm{d}}{\mathrm{d}x^{\alpha}}\left(\frac{\mathrm{d}s}{\mathrm{d}x^{\alpha}}\right)+\frac{k}{\sigma}\left[1-\frac{1}{1+\sigma s}\right]=0 \tag{28}$$

It is obvious that eq. (28) is equivalent to eq. (23) for $n = 1$. In order to study the fractal diffusion-reaction process, we introduce two variables T and E, defined, respectively:

$$T=\frac{1}{2}\left(\frac{\mathrm{d}s}{\mathrm{d}x^{\alpha}}\right)^{2} \tag{29}$$

$$E=\frac{k}{\sigma}\left[\frac{1}{\sigma}\ln(1+\sigma s)-s\right] \tag{30}$$

where T and E are, respectively, the kinetic energy and potential energy of the diffusion-reaction process.

A higher value of E means a more active reaction. The real reaction process is taken by the system between $x = 0$ and $x = L$ for which the action, eq. (25), is stationary to the first order.

Considering the boundary conditions, $s'(0)= 0$ and $s(L^\alpha) = ks_\infty$, we assume the solution has the form:

$$s = ks_\infty + c(x^{2\alpha} - L^{2\alpha}) \tag{31}$$

where c is an unknown constant. Substituting eq. (31) into eq. (25):

$$J(s) = \int_0^{L^\alpha} \left\{ 2c^2 x^{2\alpha} + \frac{k}{\sigma}\left[ks_\infty + c(x^{2\alpha} - L^{2\alpha}) - \frac{1}{\sigma}\ln(1 + \sigma ks_\infty + \sigma c(x^{2\alpha} - L^{2\alpha}) \right] \right\} \mathrm{d}x^\alpha \tag{32}$$

The stationary condition of eq. (25) can be approximated:

$$\frac{\mathrm{d}J}{\mathrm{d}c} = \int_0^{L^\alpha} \left\{ 4cx^{2\alpha} + \frac{k}{\sigma}\left[x^{2\alpha} - L^{2\alpha} - \frac{x^{2\alpha} - L^{2\alpha}}{1 + \sigma ks_\infty + \sigma c(x^{2\alpha} - L^{2\alpha})} \right] \right\} \mathrm{d}x^\alpha = 0 \tag{33}$$

or

$$\frac{4}{3}cL^{3\alpha} + \frac{k}{\sigma}\left(\frac{1}{3}L^{3\alpha} - L^{3\alpha} \right) - \frac{k}{\sigma^2 c}\left[1 - \frac{1 + \sigma ks_\infty}{(1 + \sigma ks_\infty - \sigma cL^{2\alpha})} \tan^{-1}\sqrt{\frac{\sigma c}{(1 + \sigma ks_\infty - \sigma cL^{2\alpha})}} \right] = 0 \tag{34}$$

From eq. (34), the value of c can be determined, and finally we obtained the solution:

$$s = ks_\infty + c(x^{2\alpha} - L^{2\alpha}) \tag{35}$$

where c is solved from eq. (34). Now we discuss its solution properties. By differentiating eq. (35) with respect to x:

$$\frac{\mathrm{d}s}{\mathrm{d}x} = 2\alpha cx^{2\alpha - 1} \tag{36}$$

and the diffusion flux at $x = 0$ has the following property:

$$J(0) = D\frac{\mathrm{d}s}{\mathrm{d}x}(0) = \begin{cases} 0, \alpha > \dfrac{1}{2} \\ Dc, \alpha = \dfrac{1}{2} \\ \infty, \alpha < \dfrac{1}{2} \end{cases} \tag{37}$$

Equation (37) implies when $\alpha < 1/2$, we predict an extremely fast diffusion process. To understand the threshold value of $\alpha = 1/2$, we begin with a smooth and continuous electrode, such a case predicts $\alpha = 1$, and the traditional model works well. When the electrode has porous structure, and the porosity area takes up half of the surface area, we have $\alpha = 1/2$ [18, 19]. So $\alpha < 1/2$ implies the porosity area is larger than 1/2 of the total surface area.

In case $\alpha > 3/2$:

$$J(0) = \frac{\mathrm{d}J}{\mathrm{d}x}(0) = \frac{\mathrm{d}^2 J}{\mathrm{d}x^2}(0) = \frac{\mathrm{d}^3 J}{\mathrm{d}x^3}(0) = 0 \tag{38}$$

Equation (38) implies an extremely slow diffusion process at $x = 0$. As previously discussed $\alpha < 1$ implies a porous surface, like the Menger sponge; and when $\alpha > 1$, we have a lotus-leaf-like uneven surface.

Discussion and conclusions

If a higher order approximate solution is needed, we can assume:

$$s = ks_{\infty} - a_2(x^{2\alpha} - L^{2\alpha}) + \cdots + a_n(x^{n\alpha} - L^{n\alpha}) \tag{39}$$

where a_i (i = 2, 3,...,n) are constants to be determined.

Substituting eq. (38) into eq. (32):

$$\frac{\partial J}{\partial a_i} = 0(i = 2,3,\cdots,n) \tag{40}$$

we can determine a_i (i = 2, 3,...,n).

The fractal diffusion-reaction equation can be also solved by the homotopy perturbation method, variational iteration method or Taylor series method [30-39].

This paper gives an ample evidence that the fractal variational approach to amperometric enzymatic reactions is not only simple, but also rigorous. The extremely fast or extremely slow diffusion process is elucidated by different surface structure of the electrode, a Menger sponge-like surface leads to an extremely fast diffusion process, while a lotus-leaf-like surface results in an extremely slow diffusion process. These properties are extremely helpful for an optimal design of an electrode with suitable a surface morphology to control the diffusion-reaction process.

References

[1] Zhokh, A., *et al*., Relationship between the Anomalous Diffusion and the Fractal Dimension of the Environment, *Chem. Phys*., *503* (2018), Mar., pp. 71-76
[2] Shanmugarajan, A., *et al*., Analytical Solution of Amperometric Enzymatic Reactions Based on Homotopy Perturbation Method, *Electrochimica Acta*, *56* (2011), 9, pp. 3345-3352
[3] Rahamathunissa, G., Rajendran, L., Application of He's Variational Iteration Method in Non-Linear Boundary Value Problems in Enzyme-Substrate Reaction Diffusion Processes – Part 1: The Steady-State Amperometric Response, *Journal Math. Chem*., *44* (2008), 3, pp. 849-861
[4] Abukhaled, M., Khuri, S. A., A Semi-Analytical Solution of Amperometric Enzymatic Reactions Based on Green's Functions and Fixed Point Iterative, *Journal of Electroanalytical Chemistry*, *792* (2017), Mar., pp. 66-71
[5] He, J. H., A Simple Approach to 1-D Convection-Diffusion Equation and Its Fractional Modification for E Reaction Arising in Rotating Disk Electrodes, *Journal of Electroanalytical Chemistry*, *854* (2019), Dec., 113565
[6] Li, X. X, *et al*., A Fractal Modification of the Surface Coverage Model for an Electrochemical Arsenic Sensor, *Electrochimica Acta*, *296* (2019), Feb., pp. 491-493
[7] He, J.-H., Variational Principle for the Generalized KdV-Burgers Equation with Fractal Derivatives for Shallow Water Waves, *Journal Appl. Comput. Mech*., *6* (2020), 4, pp. 735-740
[8] Li, X. J., He, J. H., Variational Multi-Scale Finite Eement Method for the Two-Phase Flow of Polymer Melt Filling Process, *International Journal of Numerical Methods for Heat and Fluid-Flow*, *30* (2019), 3, pp. 1407-1426
[9] He, J.-H., Variational Principle and Periodic Solution of the Kundu-Mukherjee-Naskar Equation, *Results in Physics*, *17* (2020), June, 103031
[10] He, J. H., Lagrange Crisis and Generalized Variational Principle for 3-D Unsteady Flow, *International Journal of Numerical Methods for Heat and Fluid-Flow*, *30* (2019), 3, pp. 1189-1196
[11] He, J. H., and Sun, C., A Variational Principle for a Thin Film Equation, *Journal of Mathematical Chemistry*, *57* (2019), 9, pp. 2075-2081
[12] He, J. H., Generalized Variational Principles for Buckling Analysis of Circular Cylinders, Acta Mechanica, *231* (2019), Dec., pp. 899-906
[13] He, J. H., A Fractal Variational Theory for 1-D Compressible Flow in a Microgravity Space, *Fractal*s, *28* (2020), 02, 2050024
[14] Malkovich, E. G., et al. Two-component model for catalyst deactivation, *Chemical Engineering Journa*l, *378* (2019), Dec., 122176

[15] Greiner, R., *et al*., Tomography Based Simulation of Reactive Flow at the Micro-Scale: Particulate Filters with Wall Integrated Catalyst, *Chemical Engineering Journal*, *378* (2019), Aug., 121919
[16] Phutke, M., *et al*., Modelling Solid-Solid Reactions: Contact-Point Approach, *Chemical Engineering Journal*, *377* (2019), SI, 120570
[17] Gonzalez-Casamachin, D. A., *et al*., Visible-Light Photocatalytic Degradation of Acid Violet 7 Dye in a Continuous Annular Reactor Using ZnO/PPy Photocatalyst: Synthesis, Characterization, Mass Transfer Effect Evaluation and Kinetic Analysis, *Chemical Engineering Journal*, *373* (2019), Oct., pp. 325-337
[18] Ain, Q. T., He, J. H., On Two-Scale Dimension and Its Applications, *Thermal Science*, *23* (2019), 3B, pp. 1707-1712
[19] He, J. H., Ji, F. Y., Two-Scale Mathematics and Fractional Calculus for Thermodynamics, *Thermal Science*, *23* (2019), 4, pp. 2131-2133
[20] He, J. H., A Tutorial Review on Fractal Spacetime and Fractional Calculus, *International Journal of Theoretical Physics*, *53* (2014), 11, pp. 3698-3718
[21] He, J. H., Fractal Calculus and Its Geometrical Explanation, *Results in Physics*, *10* (2018), Sept., pp. 272-276
[22] Wang, Q. L., *et al*., Fractal Calculus and Its Application Explanation of Biomechanism of Polar Bear, *Fractals*, *26* (2018), 6, 1850086
[23] Wang, Q. L., *et al*., Fractal Calculus and Its Application Explanation of Biomechanism of Polar Bear (Vol. 26, 1850086, 2018), *Fractals*, *27* (2019), 5, 1992001
[24] Fan, J., *et al*., Explanation of the Cell Orientation in a Nanofiber Membrane by the Geometric Potential Theory, *Results in Physics*, *15* (2019), Dec., 102537
[25] Jin, X., *et al*., Low Frequency of a Deforming Capillary Vibration, – Part 1: Mathematical Model, *Journal of Low Frequency Noise Vibration and Active Control*, *38* (2019), 3-4, pp. 1676-1680
[26] Li, X. X., He, J. H., Along the Evolution Process Kleiber's 3/4 Law Makes Way for Rubner's Surface Law: A Fractal Approach, *Fractals*, 27 (2019), 2, 1950015
[27] Tian, D., *et al*., Geometrical Potential and Nanofiber Membrane's Highly Selective Adsorption Property, *Adsorption Science and Technology*, *37* (2019), 5-6, pp. 367-388
[28] Wang, Y., *et al*., A Variational Formulation for Anisotropic Wave Traveling in a Porous Medium, *Fractals*, *27* (2019), 4, 19500476
[29] Wang, K. L., et al., A Remark on Wang's Fractal Variational Principle, *Fractals*, *27* (2019), 08, 1950134
[30] Yu, D. N., *et al*., Homotopy Perturbation Method with an Auxiliary Parameter for Non-Linear Oscillators, *Journal of Low Frequency Noise Vibration and Active Control*, *38* (2019), 3-4, pp. 1540-1554
[31] Ren, Z. F., *et al.*, He's Multiple Scales Method for Non-Linear Vibrations, *Journal of Low Frequency Noise Vibration and Active Control*, *38* (2019), 3-4, pp. 1708-1712
[32] Liu, Z. J., *et al*., Hybridization of Homotopy Perturbation Method and Laplace Transformation for the Partial Differential Equations, *Thermal Science*, *21* (2017), 4, pp. 1843-1846
[33] Adamu, M. Y., Ogenyi, P., New Approach to Parameterized Homotopy Perturbation Method, *Thermal Science*, *22* (2018), 4, pp. 1865-1870
[34] Ban, T., Cui, R. Q. , He's Homotopy Perturbation Method for Solving Time-Fractional Swift-Hohenerg Equation, *Thermal Science*, *22* (2018), 4, pp. 1601-1605
[35] Anjum, N., He, J. H., Laplace Transform: Making the Variational Iteration Method Easier, *Applied Mathematics Letters*, *92* (2019), June, pp. 134-138
[36] He, J. H., Some Asymptotic Methods for Strongly Non-Linear Equations, *International Journal of Modern Physics B*, *13* (2006), 20, pp. 1141-1199
[37] He, J. H., The Simpler, the Better: Analytical Methods for Non-Linear Oscillators and Fractional Oscillators, *Journal of Low Frequency Noise Vibration and Active Control*, *38* (2019), 3-4, pp. 1252-1260
[38] He, J. H., Ji, F. Y., Taylor Series Solution for Lane-Emden Equation, *Journal of Mathematical Chemistry*, *57* (2019), 8, pp. 1932-1934
[39] He, C. H., et al., Taylor Series Solution for Fractal Bratu-Type Equation Arising in Electrospinning Process, *Fractals*, *28* (2020), 1, 2050011

Paper submitted: December 12, 2019
Paper revised: June 12, 2020
Paper accepted: June 12, 2020