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Calculation of the radiative properties of photosynthetic microorganisms

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A B S T R A C T

A generic methodological chain for the predictive calculation of the light-scattering and absorption properties of photosynthetic microorganisms within the visible spectrum is presented here. This methodology has been developed in order to provide the radiative properties needed for the analysis of radiative transfer within photobioreactor processes, with a view to enable their optimization for large-scale sustainable production of chemicals for energy and chemistry. It gathers an electromagnetic model of light-particle interaction along with detailed and validated protocols for the determination of input parameters: morphological and structural characteristics of the studied micro-organisms as well as their photosynthetic-pigment content. The microorganisms are described as homogeneous equivalent-particles whose shape and size distribution is characterized by image analysis. The imaginary part of their refractive index is obtained thanks to a new and quite extended database of the *in vivo* absorption spectra of photosynthetic pigments (that is made available to the reader). The real part of the refractive index is then calculated by using the singly subtractive Kramers–Krönig approximation, for which the anchor point is determined with the Bruggeman mixing rule, based on the volume fraction of the microorganism internal-structures and their refractive indices (extracted from a database). Afterwards, the radiative properties are estimated using the Schiff approximation for spheroidal or cylindrical particles, as a first step toward the description of the complexity and diversity of the shapes encountered within the microbial world. Finally, these predictive results are confronted to experimental normal-hemispherical transmittance spectra for validation. This entire procedure is implemented for *Rhodospirillum rubrum*, *Arthrospira platensis* and *Chlamydomonas reinhardtii*, each representative of the main three kinds of photosynthetic microorganisms, *i.e.* respectively photosynthetic bacteria, cyanobacteria and eukaryotic microalgae. The obtained results are in very good agreement with the experimental measurements when the shape of the microorganisms is well described (in comparison to the standard

Acronyms: DDA, Discrete-Dipole Approximation; FDTD, Finite Difference Time Domain; OD, Optical Density; PAR, Photosynthetically Active Radiation

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volume-equivalent sphere approximation). As a main perspective, the consideration of the helical shape of *Arthrospira platensis* appears to be a key to an accurate estimation of its radiative properties. On the whole, the presented methodological chain also appears of great interest for other scientific communities such as atmospheric science, oceanography, astrophysics and engineering.

1. Introduction

Photosynthesis engineering is nowadays recognized as a possible alternative to the exhaustion of fossil resources and global warming. Indeed, it enables both the mitigation of CO_2 emission and the sustainable synthesis of chemicals for energy (biomass, biofuels) as well as for chemistry (raw or high-value green products). Nevertheless, first the development of new and efficient technologies like photobioreactors devoted to the photosynthetic-microorganism cultivation with thermodynamic efficiencies approaching at least 10% is required. Such original future designs, or more basically the optimization of existing concepts, are only feasible if predictive knowledge-models of these processes are developed on a strong physical basis, giving them sufficient genericity for industrial purposes (simulation, sizing, scale-up, optimization, model-based predictive control, *etc.*). Assuming a proper mixing and that all physiological needs are maintained at their optimal conditions (pH, temperature, dissolved CO_2, minerals), it has been clearly demonstrated in the past decades that photobioreactors are mainly governed by radiant light transfer inside the culture volume, determining the kinetic rates, energetic yields, biomass composition and pigment contents [1–8]. Accurate knowledge of the radiation fields is therefore required prior to any analysis of photosynthesis engineering, which is only feasible by properly solving the radiative transfer equation for any boundary condition, *i.e.* any photobioreactor design [1,9,8,10–12]. This equation contains three parameters, respectively the absorption and scattering coefficients together with the scattering phase-function, which must be known with accuracy if the resulting radiation-fields are envisaged to formulate a kinetic and energetic model of the reactor. Therefore, any radiative analysis of photobioreactors starts with the determination of the absorption and scattering properties of the involved photosynthetic-microorganisms. Yet this question is not trivial, and to the best of our knowledge, no available database provides the adequate spectral and angular information needed, even for the strains of microalgae that are currently widely cultivated. Obtaining these properties involves either highly specialized experiments [13–17], or the construction of a model implying the resolution of Maxwell's equations for particles with types of heterogeneities, sizes and shapes for which the usual numerical methods such as Lorenz–Mie, T-Matrix, FDTD, DDA, *etc.* [18,19] are yet impracticable in most cases. Within the international literature dedicated to the study of photobioreactors, L. Pilon's research group (University of California, Los Angeles) is the only one, to the best of our knowledge, that focuses on the experimental determination of the radiative properties of photosynthetic microorganisms, through single scattering experiments including the measurement of the angular distribution of the scattered power [15,16,8]. The present paper addresses the electromagnetic modelling approach, and we hereafter present a methodological chain inherited from the expertise of the oceanography community, and more broadly, of all the communities that are confronted with the wave–particle interaction problem, in either atmospheric sciences, astrophysics or engineering. In this wide context, our concern is to account not only for the specificity of photosynthetic microorganisms, but also for the analysis and optimization requirements of photobioreactor-engineering.

The above-mentioned theoretical and experimental issues have gradually developed during the past 15 years, but yet they have remained overwhelmingly open-ended. In the light of this gap, numerous research groups have no alternatives but to make use of the empirical tools developed by the community over the past 20 years [20–25,10]. These tools are based on radically different modelling approaches from the one that we develop hereafter. The spacial dependence of radiative observable is indeed fixed *a priori* according to laws inspired by radiation physics (Beer–Lambert extinction law, hyperbolic laws, *etc.*) that involve a set of free parameters to be adjusted, on a case by case basis, so as to conform to available experimental data. Of course these empirical approaches reach their limits when considering diverse operating modes of photobioreactors, due to the necessity to adjust the parameters for each of the studied situations (*e.g.* reactor geometry, incident light flux and radiation frequency), or simply when considering a process with complex geometry (experimental data then being difficult to obtain). These are the constraints that have led us to identify the need for a predictive methodology, based on limited and "easily" accessible experimental parameters, allowing us to grasp the microorganisms' variability, from one species to another and as a function of the culture conditions (in particular as a function of the illuminating conditions) [26,11,27]. Within this framework, the purpose of the present paper is to gather and extend our know-how in order to design a full methodological chain ranging from the determination of the input parameters of our model to the validation of its results.

In the following, we make use of several methods that are furthermore described, independently of each other, within the literature dedicated to light scattering by small particles [28–31]. The very general problem of modelling radiative properties is fully posed in these works, the formalism of which is here adopted with the conventions