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Kinetics analysis and quantitative calculations for the successive radioactive decay process

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Abstract

The general radioactive decay kinetics equations with branching were developed and the analytical solutions were derived by Laplace transform method. The time dependence of all the nuclide concentrations can be easily obtained by applying the equations to any known radioactive decay series. Taking the example of thorium radioactive decay series, the concentration evolution over time of various nuclide members in the family has been given by the quantitative numerical calculations with a computer. The method can be applied to the quantitative prediction and analysis for the daughter nuclides in the successive decay with branching of the complicated radioactive processes, such as the natural radioactive decay series, nuclear reactor, nuclear waste disposal, nuclear spallation, synthesis and identification of superheavy nuclides, radioactive ion beam physics and chemistry, etc.

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1. Introduction

Among the thousands of known nuclides, the most are unstable, i.e. radioactive. The initial radioactive nuclide is called the parent nuclide and the new one obtained from decay the daughter. If the daughter nuclide is also unstable, it will decay again and, in this way, the process continues until it becomes a final stable nuclide. As we know, many radioactive decay is a successive process. A daughter nuclide is often the parent of another decay process. Therefore, the concentration change of nuclides is very complicated for a continuous successive decay process. For any specified nuclide, its decay is random. However, its decay rate can be determined in the statistical sense. In nature, there are four typical radioactive decay series based on the parent nuclide 232 Th, 237 Np, 238 U, and 235 U respectively. Mass numbers of their family members are in the 4n, 4n + 1, 4n + 2, 4n + 3 categories, respectively, where *n* is an integer. Kinetics study on radioactive decay is of important scientific significance and application value for the nuclear reactor design, nuclear waste disposal, synthesis and identification of superheavy nuclides, accelerator driven system design, etc.

Early in 1910, Bateman [1] developed a set of kinetics equations for a linear radioactive decay chain, in which every parent has only a single child. The analytical solutions of the Bateman equations, i.e. the concentration evolution over time of nuclide numbers undergoing serial decay, were obtained by the Laplace transform method. However, in the series decay process, some nuclides may undergo two or more different decay reactions and generate two or more child nuclides, which is called branching decay. As pointed out by some authors [2,3], Bateman's work did not consider the complicated branching decay situations, and he originally only provided solutions for the systems with less than five nuclides. In addition, most classic nuclear physics textbooks rarely discussed analytic solutions of decay chains with more than three nuclides [4,5] or with branches [6,7].

In addition, Bateman's solutions are only applicable to systems in which all decay constants are different. Otherwise infinity will occur (zero in denominator). Thus, for a radioactive decay process with branching decay, the decay kinetics equations based on a decay mode without branching should be revised with care. Under these circumstances, great efforts have been made to solve and revise the Batman equations since its inception. A short introduction for obtaining the general solution was demonstrated by some authors using the Laplace transform method [3,8]. In order to exclude the infinity possibility, Cetnar [9] derived the general solution for the decay equation of linear chain. Algebraic approach to solve Bateman equations were also presented by many authors [10–15]. Yuan and Kernan [16] introduced the definition of a more generic class of radioactive decay chains—the "exit-only" decay chain. For such decay chains, they derived explicit solutions for decay equations under given initial-value conditions. Furthermore, the E-factor function method [17], the recursive method [18] and the mode method [19] were developed, as well. Strenge [20] presented a general solution to first-order compartmental models, which can be applied to any systems involving physical transfers from the medium and radioactive chain decay with branching in the medium.

For the purpose of calculation skills, Laplace transform method has a great advantage. It relies on a Laplace transform and its subsequent inverse transform using a path integral in the complex plane. The algebraic approach derived the solution on the basis of the eigenvalues and the correspondent eigenvectors of the decay constants matrix. In this work, by Laplace transform method, we try to derive the explicit solutions to the kinetics equations of radioactive decay series with branching and apply it to the known natural radioactive decay series. The numerical calculations are taken to verify reliability of the method.

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