

keV ion irradiation assisted prebiotic synthesis of oligopeptide in the solar system

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Abstract

In this paper, keV ion assisted peptide formation from amino acid monomers is studied. To investigate the possible role of low energy ion irradiation in prebiotic synthesis of oligopeptides, we applied keV Ar⁺ and N⁺ ion beams to solid state phenylalanine. After ion irradiation, it was found that phenylalanine dipeptide was formed and the optimal pH value for peptide formation was 8–9. A radical mechanism was also deduced for our observations. In view of the ubiquitous existence of keV ions in early space and on the early Earth, we suggest that low energy charged particles might have played a role in chemical evolution in the solar system.

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1. Introduction

According to chemical evolution theory, the condensation of amino acids to form peptides in the prebiotic environment is believed to have been a vital step in the chemical origin and evolution of life (Lemmon, 1970). The general problem of peptide formation from amino acids is the thermodynamically and kinetically unfavorable process of dehydration (Meggy, 1956). To overcome the thermodynamic barrier, a variety of catalysts and external energy are necessary: a catalyst can decrease the activation energy of chemical reaction and the external energy may promote cross-condensation or self-condensation reaction of amino acids.

Hypothetical possibilities contributing to the condensation of amino acids under prebiotic conditions include melt processes (Fox and Harada, 1958, 1960), condensation reagents mediated peptide formation (Steinman et al., 1965; Ponnampertuma and Peterson, 1965; Ehler and Orgel, 1976; Ferris et al., 1996), thermal condensation on the sur-

face of minerals (Lahav et al., 1978; Rao et al., 1980; Bujdák and Rode, 1999), salt-induced peptide formation (SIPF) (Rode and Schwendinger, 1990; Rode, 1999), CO activated condensation on (Ni,Fe)S surface (Huber and Wächtershäuser, 1998; Huber et al., 2003), peptide formation in a simulated submarine hydrothermal system (Imai et al., 1999; Alargov et al., 2002), and carbonyl sulfide (COS) induced coupling (Leman et al., 2004). However, some of these hypothetical conditions could not have existed in primordial times, (Rode, 1999), and some do not work efficiently; moreover, any suggested mechanism needs to be demonstrated to be effective under simulated early Earth conditions.

Exogenous delivery of organic matter to the Earth is believed to be an important item of the inventory for the origins of life, as well as endogenous production and impact-shock synthesis (Chyba and Sagan, 1992). Over 80 distinct amino acids have been observed in a number of carbonaceous chondrite meteorites of different classes; the indigenous nature of these amino acids is confirmed by the presence of non-terrestrial isotopic ratios (Kvenvolden et al., 1970; Cronin and Pizzarello, 1983; Epstein et al., 1987; Llorca, 2004; Glavin et al., 2006). Many

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authors suggested that small bodies in the solar system such as comets, carbonaceous chondrites and interplanetary dust particles are rich in organics including amino acids, and irradiated by ultraviolet photons and cosmic particles which are the main energy sources for abiogenic synthesis of biomolecules in space (Lemmon, 1970; Simakov et al., 1996, 1997; Kobayashi et al., 2001). They bombarded solid state amino acids with UV photons and high energy protons (6.6 MeV), and dipeptides were identified (Simakov et al., 1996, 1997).

Actually, energetic ions with energy ranging from keV to MeV, such as cosmic rays, magnetospheric ions, solar flares, and solar wind particles, are ubiquitous in space (Brucato et al., 1997; Strazzulla et al., 2003). Due to their impact, some chemical reactions take place on the refractory and/or frozen surface of celestial bodies. These processes could have played a key role in the early chemical evolution (Strazzulla, 1997; Strazzulla and Palumbo, 2001). In this paper, we use keV ions (unreactive Ar^+ and reactive N^+) to irradiate solid state phenylalanine (Phe). The aim of this work is to clarify the role of low energy ions in the prebiotic peptide formation.

2. Materials and methods

2.1. Materials

L-Phenylalanine (L-Phe) and its dipeptide (Phe-Phe) were purchased from Sigma Company and used without further purification. Triple-distilled water was prepared by double distillation of distilled water. All other chemicals were obtained in analytical grade, with the exception of acetonitrile (CAN) and trifluoroacetic acid (TFA) that were of chromatographic grade.

2.2. Experimental methods

2.2.1. Sample preparation

L-Phenylalanine was dissolved in triple-distilled water to give solutions with a concentration of 0.1 M at room temperature. The pH values of the solutions were adjusted as required using dilute solutions of HCl and NaOH. The solutions were put into flat-bottomed glass culture dishes with inner radius of 5.8 cm, and then dried at low temperature ($<50^\circ\text{C}$) to form solid thin films with mass thickness of 4.7 mg/cm^2 .

2.2.1.1. Irradiation. Ion beams were produced by the ion implanter in our laboratory. The thin films of L-phenylalanine were bombarded by pulsed nitrogen or argon ions at the energy of 30 keV, with doses of 10^{15} – 10^{16} cm^{-2} , pulse time 30 s, pulse interval 5 s, pulse dose about $2.5 \times 10^{15}\text{ cm}^{-2}$. Each experiment at different pH and different dose levels was repeated three times. During ion implantation, the sample was left on a backing and cooled with water to keep the temperature of the sample lower than 50°C .

2.2.1.2. HPLC analyses. After irradiation of the solid films, resulting products were redissolved in triple-distilled water and analyzed using high performance liquid chromatograph (HPLC, WatersTM). A special reverse phase column (VydacTM 218TP5415, C18, $5\ \mu\text{m}$, $150 \times 4.6\text{ mm}$) for peptide analysis was used. HPLC conditions employed were as follows: mobile phase, $\text{ACN:H}_2\text{O:TFA} = 20:80:0.08$, v/v; flow rate, 1.0 mL/min ; UV detection, 258 nm .

3. Results and discussion

3.1. Phe dipeptide formation

When the irradiated solid films of Phe were redissolved in triple-distilled water and were applied to HPLC detection, Phe dipeptide was identified in the aqueous solutions. As shown in Fig. 1(b) and (c), the separation profile of the irradiated sample had a 6.6 min peak which was identical with the standard Phe dipeptide, while the control experiment without irradiation did not show any trace of oligopeptide (Fig. 1(a)). Using the HPLC detection method, therefore, we demonstrate the synthesis of Phe dipeptide from phenylalanine monomer after keV ion implantation.

In discussing the possible mechanism of peptide formation, it should be taken into account that the peptide is formed through radical-based condensation reaction. Ion irradiation of amino acids can produce radicals (Huang et al., 1998). In its quest to achieve stability, the active radical spontaneously tries to either pass on the extra electron to its nearest neighbor, or grab an electron from the neighbor to make up a pair to revolve around its own nucleus. Thus the radical precursors generated by ion irradiation can recombine with another to become a totally new molecule, setting off a peptide formation reaction.

The model chemistry based on radical–radical or radical–molecule reaction would explain the existence of dipep-

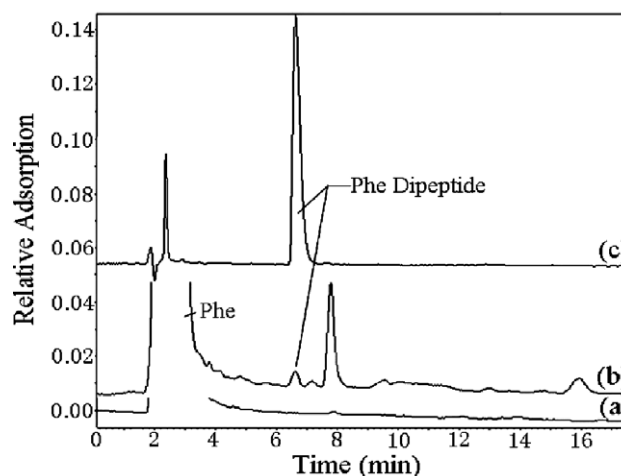


Fig. 1. HPLC elution profiles of a control Phe sample, an irradiated sample and the standard Phe dipeptide (Traces a, b, and c, respectively). Irradiation conditions: $\text{pH} = 9.0$, 30 keV Ar^+ , $2.5 \times 10^{16}\text{ ions/cm}^2$.

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