

 MLF Experimental Report	提出日 Date of Report 2013/7/22
課題番号 Project No. 2013A0145 実験課題名 Title of experiment Hydration effect on electron transfer process in cytochrome <i>c</i> and DNA probed by muon labelling method 実験責任者名 Name of principal investigator Yoko Sugawara 所属 Affiliation Kitasato University	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) Muon D1 実施日 Date of Experiment 2013/5/7 9:00 – 2013/5/10 21:00 (The beam supply was stopped during 5/8 9:00 a.m. to 5/9 4:30 a.m.)

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)
 Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form. Cytochrome <i>c</i> (104 amino acid residues with a heme from horse heart), whose water contents were adjusted to approximately 5 % (a dry sample) and 20% (a wet sample).
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2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。) Experimental method and results. If you failed to conduct experiment as planned, please describe reasons. In order to investigate a hydration effect on the electron transfer process of cytochrome <i>c</i> and nucleic acid, we intended to measure the μ SR time spectra of the following three samples. 1. Dried sample of cytochrome <i>c</i> (water content approximately 5 %) 2. Wetted sample of cytochrome <i>c</i> (water content approximately 20 %) 3. Dried sample of deoxyribonucleic acid (DNA) At first, we checked whether the single or the double pulse was suitable for the measurements. In order to obtain information just after a pulse radiation by removing the second pulse arriving at 600 ns after the first pulse, we concluded that the use of the single pulse was suitable admitting 50 % counting rate. After that, we carried out following measurements. 1) Measurements of longitudinal μ SR time spectra of the wet sample of cytochrome <i>c</i> , under the longitudinal fields of 0, 0.1, 0.5, 1.0 and 1.4 kG, at 300 K, 250K, 200K and 150K. 2) Measurements of longitudinal μ SR time spectra of the dry sample of cytochrome <i>c</i> , under the longitudinal fields of 0, 0.1, 0.5, 1.0 and 1.4 kG, at 300 K, 250 K and 200 K.

2. 実験方法及び結果(つづき) Experimental method and results (continued)

A significant difference between these samples was found in temperature (T) dependence of the zero field relaxation function (Figure 1). Following zero-field part of the Risch-Kehr theory (R. Risch, K.W. Kehr, Phys. Rev. B 46 (1992) 5246.), the observed result suggests a T-dependent change of electron transport: slow (rapid) at low (high) temperature in the wet sample, while T-independent rapid transport in the dry sample. We plotted the longitudinal relaxation function for each μ SR time spectrum against longitudinal field (B) (Figure 2). The result was compared to the prediction of the Risch-Kehr theory: relaxation parameter Γ of the Risch-Kehr relaxation function is proportional to $1/B$, when the electron brought-in by the μ^+ takes one-dimensional motion along the molecular chain. A relaxation parameter Γ in the wet sample shows only weak field dependence up to 1000G at 300 K, while that in the dry sample has apparent an inverse dependence on magnetic field at all T. The results may suggest an existence of inter-chain electron diffusion through the hydration water in the wet cytochrome c at high T, while the intra-chain electron transfer is more dominant in the dry sample at all T.

We are planning a series of experiments on deuterides of wet cytochrome c in order to confirm a non-contribution of nuclear dipolar relaxation on the observed hydration effect. The experiments on the samples of deoxyribonucleic acid, which could be not measured during this beam time, will also be scheduled.

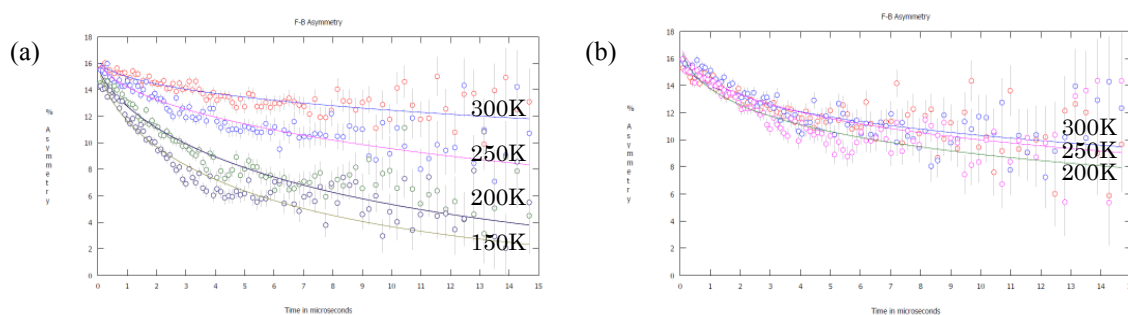


Fig.1 Temperature dependence of the zero field relaxation function of cytochrome c.

(a) Wetted sample (water content approximately 20%) (b) Dried sample (water content approximately 5%)

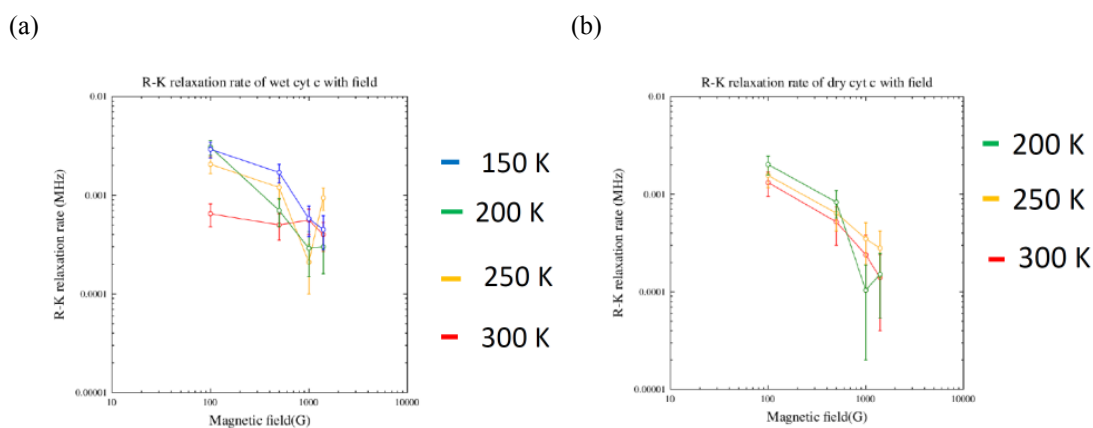


Fig.2 Magnetic field dependence of the R-K relaxation rates of cytochrome c

(a) Wetted sample (water content approximately 20%) (b) Dried sample (water content approximately 5%)