Biochimica et Biophysica Acta 1804 (2010) 1–2

Contents lists available at ScienceDirect

Biochimica et Biophysica Acta

journal homepage: www.elsevier.com/locate/bbapap

Preface

The Protein-Water Energy Seascape

Protein molecules and liquid water are very different systems both from a structural and a dynamic point of view. Proteins are long range ordered, but its local molecular motions are restricted by covalent bonds, which makes it solid-like. The dynamics of such a system is properly represented by barrier crossing within a well defined energy landscape. Water in contrast is short-range ordered, but long range diffusion allows it to flow, the essence of the liquid state. The energy landscape is thus fluctuating and motion is determined by collective reorganization of molecules forming transient holes, the landscape turns into a seascape. At the protein-water interface mutual interactions modify the properties of both systems. The protein landscape is modulated in time by density fluctuations in the liquid phase and interfacial water is transiently immobilized by rigid protein structure.

The physical phenomenon of protein hydration is to some extent elusive, since there is no rigid shell of water around a protein molecule. Instead there is a fluctuating cloud which is thermodynamically and dynamically different from bulk water. A "hydrodynamic" definition of hydration water is to evaluate, how many water molecules near the protein surface have their displacement vector along the trajectory of the protein at any instant of time. This is determined by the strength of interaction with the protein and, hence, by the ratio of the time that is spent "attached" to the protein to the time spent in the bulk phase. The sum of the fractional contributions of all water molecules is the measured hydration. It is the mass of water, which migrates with the protein at any instant of time. When divided by the molecular weight of water, this yields the average number of molecules, which interact with the protein [1].

An operational definition is to determine the amount of non-freezable water at low temperatures, which depends however on the cooling-rate if crystallization occurs. Below about 0.4 g water per g protein, crystallization is hindered. The hydration shell can be supercooled until a glass transition interferes at 170 K. In such hydrated protein powders, the water molecules are permanently in contact with the protein surface, while protein translation and rotation is suppressed. Such surface-attached water molecules are still highly mobile, performing long-range diffusion, which is a property of the liquid state. The translational diffusion is arrested at the glass transition [2]. This dramatic slowing down of molecular motions of water is a useful approach to the coupling between water and protein dynamics.

With neutron scattering experiments with hydrated myoglobin and lysozyme a dynamical transition was observed in terms of two spectral components [3]: i) a fast local process, termed β-process, which did not slow down with decreasing temperature, only the amplitude was diminished. It was tentatively assigned to fast protein-water hydrogen bond fluctuations. ii) a slower water-coupled relaxation was identified, the α-process, which slows down with decreasing temperature in parallel with the motion of hydration water. The α-process is also present in hydration water. It represents the first step of translational diffusion of water along the protein surface and defines a surface viscosity.

The surface viscosity is the central parameter, characterizing the dynamic interactions between protein residues at the surface and the solvation shell. W. Doster shows in his contribution in this special issue, that functional properties like ligand exchange depend on the viscosity near the protein surface, which can be different from the bulk due to preferential hydration [1].

This special issue of BBA - Proteins and Proteomics on "Biomolecular Dynamics" summarizes contributions given at a workshop held in Feldafing near Munich, Germany, in September 2008 on "Biomolecular Dynamics and Protein-Water Interactions – A Neutron Scattering Workshop". Neutron scattering can probe fast protein-water dynamics on a pico- to nano-second time scale using various neutron spectroscopic methods. This information has been compared with results obtained with other methods such as dielectric relaxation and NMR spectroscopy in this workshop.

Hence, A. Sokolov et al. presents a description of the glass transition in hydrated proteins using neutron scattering, while H. Jansson et al. reviews in their contribution results measured by dielectric spectroscopy and calorimetry. M. Kataoka and co-workers shows the effect of conformational states on the protein dynamical transition with neutron scattering. G. Kearley et al. looks at the dynamical transition in large globular proteins with the same technique. Studies using NMR spectroscopy on water and protein dynamics are presented by M. Vogel and colleagues. S. Magazu et al. characterizes protein motion in myoglobin applying self-distribution-function procedure on neutron data. A comparison of neutron experiments and molecular dynamics simulations is given by G. Kneller on the self-dynamics of proteins under hydrostatic pressure. Results of neutron scattering experiments on protein dynamics under pressure are also given by A. Filabozzi et al. The diffusion of proteins in crowded solution is tackled by F. Schreiber et al. applying neutron back-scattering and spin-echo spectroscopy. An approach to separate coherent and incoherent scattering in neutron scattering experiments of protein samples by polarization analysis is presented by A. Gaspar et al. Finally the possibility of time resolved quasi-elastic neutron scattering experiments are shown in the contribution by J. Pieper.

The contributions collected reflect the current state of available experimental techniques and theoretical approaches to understand protein-water dynamics. In particular progress in simulation and better access to high flux neutron spectrometers and comparison with results by complementary techniques will lead to a better understanding of the underlying processes driving protein motions, protein folding and unfolding and protein stability essential to life.

1570-9639/$ – see front matter
doi:10.1016/j.bbapap.2009.11.003

References

[1] S.N. Timasheff, Protein hydration, thermodynamic binding and preferential hydration, Biochem 41 (2002) 13473.
[2] Y.A. Goddard, J.P. Korb, R.G. Bryant, Structural and Dynamical Examination of the Low-Temperature Glass Transition in Serum Albumin, Biophys. J. 91 (2006) 3841.
[3] W. Doster, S. Cusack, W. Petry, Dynamical transition in hydrated myoglobin revealed by inelastic neutron scattering, Nature 337 (1989) 754.

Wolfgang Doster studied physics at the Universities in Stuttgart and Ulm (Germany). He received his diplom in 1975 with work in polymer physics on NMR spectra of de Genne's reptation model. For his doctorial thesis he performed dynamic light scattering experiments on the folding-unfolding transition of oligomeric proteins and ribosomes at the Max Planck Institut in Dortmund under the direction of Benno Hess. He received his PhD in 1979. After this he performed low temperature studies of protein dynamics as a postdoc with Hans Frauenfelder at the University of Illinois during 1981-1983. At 1984 he assumed a tenure track position at the Technical University Munich focusing on protein-solvent interactions and fast protein dynamics with neutron scattering in comparison with molecular dynamic simulations. In 1989 he introduced the concept of a “dynamical transition in proteins”, which is triggered by the liquid-glass transition of the hydration shell. Presently he is interested in high pressure studies of proteins, protein diffusion in crowded media and inside biological cells and applications of neutron scattering to biological systems in general.

Thomas Gutberlet studied chemistry at the Freie Universität in Berlin (Germany). He received his diploma in 1989. In his PhD work he studied bacterial model membrane systems with diffractin techniques in the group of Hans Bradaczek at the Institute of Cristallography in Berlin. He received his PhD in 1995. Between 1995-1998 he was PostDoc in the group of Gotthard Klose at the University of Leipzig, where he continued to study biomembrane model systems by x-ray and neutron diffraction. In 1999 he moved to the Hahn-Meitner-Institut in Berlin working with Ferenc Mezei on neutron instrumentation for the European Spallation Source project. In 2002 he joined the Paul Scherrer Institut in Switzerland, working as instrument scientist. He continued to study structure and dynamics of model membrane systems in particular in interaction with interfaces. In 2007 he moved to the Forschungszentrum Jülich GmbH to manage the user program of the Jülich Centre for Neutron Science at the FRM II in Garching. He has been co-editor of volumes on lipid bilayers, neutron spin echo spectroscopy and neutron scattering in biology.

Wolfgang Doster
Technical University Munich,
Physics Department E 13, James Franckstrasse,
D-85748 Garching, Germany

Thomas Gutberlet
Forschungszentrum Jülich GmbH,
Jülich Centre for Neutron Science at FRM II Lichtenbergstr. 1,
85747 Garching, Germany