Study of Molecular-Colloidal Solutions of C_{60} Fullerenes in Water by Small-Angle Neutron Scattering

M.V.Avdeev, V.L.Aksenov
Frank Laboratory of Neutron Physics,
Joint Institute for Nuclear Research,
141980, Dubna, Moscow Region, Russia

G.V.Andrievsky, L.I.Derevyanchenko, V.K.Klochkov
Institute for Therapy of Ukrainian AMS,
Postyshev ave. 2-a, 61039, Kharkov, Ukraine

A.A.Khokhryakov
Kyiv Taras Shevchenko National University,
Volodymyrska str. 64, 01033, Kyiv, Ukraine

Since the discovery of fullerenes (1985) their biological activity was a question of particular interest. As a consequence, biological effects of fullerenes were actively studied for the last 10 years. This kind of investigations is connected mainly with the so-called “water-soluble” fullerenes — hydrophilic chemical derivatives of fullerenes. This approach was determined by a commonly accepted opinion that fullerenes are typical hydrophobic molecules and cannot form true solutions in water. However, starting from 1994 Andrievsky G.V and co-workers developed [1,2] a method of fabricating aqueous molecular-colloidal solutions of C_{60} fullerene (C_{60}FWS) with the properties of both true and colloidal solutions simultaneously. This method is based on the transferring of fullerene from an organic solution into the aqueous phase with the help of ultrasonic treatment. The main constituent of the resulting solution is the hydrated fullerene (HyFn = $C_{60}\cdot\{H_2O\}_n$), which can form spherical clusters, $(C_{60}\cdot\{H_2O\}_n)_m$, of different sizes depending on the fullerene concentration. It was found that HyFn are powerful antioxidants and have a significant therapeutic effect when treating experimental pathologies [3,4]. The mechanism of this effect is assumed to be quite different from that of other antioxidants.

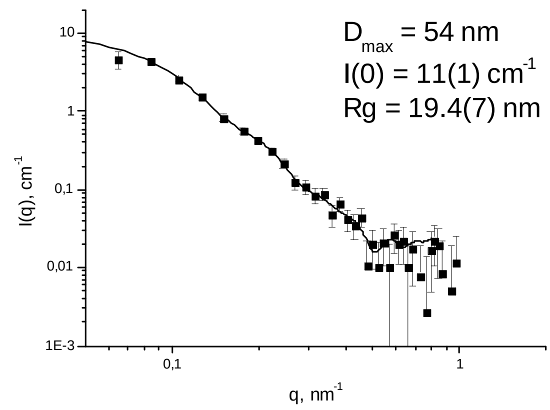
In the present work the small-angle neutron scattering (SANS) is applied to study the structure of C_{60}FWS which was produced without resort to any solubilizers and chemical modification. In the studied samples of C_{60}FWS the C_{60} concentration was from 0.002 to 0.2 mM (0.0014-0.14 mg/ml).

The SANS experiments showed (Fig.1) that these systems are highly polydisperse in a wide scale interval up to 50 nm, which testifies the electron microscopy data [5]. The attempts to obtain the stable size distribution function of aggregates using the uniform spherical form-factors fail. Along with it the contrast variation (Fig.2) performed by diluting C_{60}FWS with heavy water points to the presence of a component in the aggregates, which is different from fullerenes. A number of hypotheses about the origin of this component, in particular the formation of a special hydration shell around the fullerenes, are discussed. Also, the structural changes determined from the scattering curves after the coagulation which takes place in the solutions on addition of salts are analyzed.

The work was carried out with the support of the Russian Ministry of Industry, Science and Technologies, state contract №40.012.1.1.1148.

[1] G.V. Andrievsky et al., J. Chem. Soc., Chem. Commun., 12 (1995) 1281.
[2] G.V. Andrievsky et al., Chem. Phys. Lett., 364 (2002) 8; and UA patent N29540 of 11/15/2000.
[3] A.D. Roslyakov, et.al., Zh. Akad. Med. Nauk Ukrainy. 5 (1999) 338 (in Russian).
[4] G.V. Andrievsky, I.S. Burenin. *On Medicinal And Preventive Efficacy Of Small Doses Of Hydrated C_{60} Fullerenes At Cancer Pathologies*. Chemistry Preprint Server: http://preprint.chemweb.com/medichem/0206001. ChemWeb.com News Bulletin , 11 June, 2002.
[5] G.V. Andrievsky et al., Chem. Phys. Lett. 300 (1999) 392

a)



b)

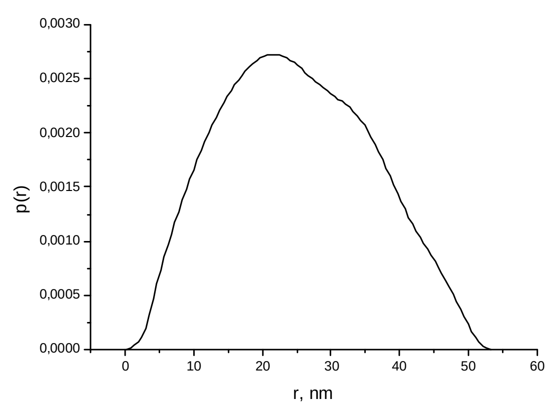


Fig.1. Typical small-angle neutron scattering curve (*a*) and the pare distance distribution function (b) for C_{60}FWS, c = 0.192 mM, T = 20°C.

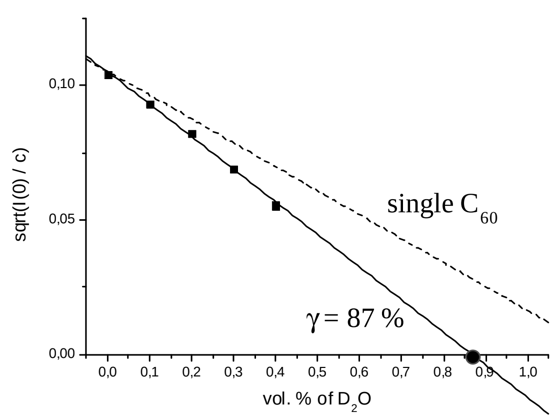


Fig. 2. Contrast variation for C_{60}FWS, c = 0.192 mM, T = 20°C; γ is the match point of the system corresponding to relative content of D_2O when the scattering from the aggregates disappears. Dashed line corresponds to behavior of the contrast as if fullerenes would be single.