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Structure and dynamics of hydrogels and organogels: An NMR spectroscopy approach

Yury E. Shapiro*

Mina and Everard Goodman Department of Life Sciences, Bar-Ilan University, Ramat-Gan 52900, Israel

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ABSTRACT

Hydrogels and organogels are semi-solid systems, in which a liquid phase is immobilized by a three-dimensional network composed of self-assembled, intertwined polymer/gelator fibers. Investigations pertaining to these systems have only picked up speed in the last few decades. Consequently, many burning questions regarding these systems, such as the specific molecular requirements guaranteeing gelation, still await definite answers. Nonetheless, the application of different hydrogels and organogels to various areas of interest, i.e., as drug delivery devices, has been quick to follow their discoveries.

The use of NMR spectroscopy for the characterization of polymer hydrogels and organogels has recently seen enormous growth. The NMR measurements involving magic angle spinning (MAS) in the solid-state NMR, spin relaxation times, nuclear Overhauser enhancements (NOE), or multiple-quantum (MQ) spectroscopy, the pulse field gradient (PFG) technique and magnetic resonance imaging (MRI) allow obtaining the detailed information on morphology, molecular organization, specific interactions and internal mobility of constituents.

This review aims at providing a global view and capabilities all of these NMR methods in comprehensive studies of hydrogels and organogels, with special emphasis on the interplay between the morphology and molecular mobility of constituents and the intermolecular interactions.

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* Tel.: +972 3 5317926; fax: +972 3 7384058.
E-mail address: shapiro@nmrsgi4.ls.biu.ac.il

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1. Introduction

A recent issue in supramolecular chemistry of smart materials is the focus on the organization of monomer and polymer species into desired superstructures. Hydrogels and organogels belong to these materials and are characterized by more than one length scale through crosslinks formed by covalent bonds or physical (hydrogen bonding, solvophobic, charge transfer and van der Waals) interactions.

NMR spectroscopy in combination with FTIR, X-ray, electron microscopy and others, is rewarding in study of morphology, molecular structure and component dynamics of gel networks. For example, the chemical shifts and intensities of peaks in the NMR spectra allow structural quantities such as polymer composition, microtacticity, sequence distribution, branching, crosslinking and molecular weight to be measured [1–4], while the more sophisticated experiments, i.e., magic angle spinning (MAS), measurements of spin relaxation times, nuclear Overhauser enhancements (NOE), or multiple-quantum (MQ) spectroscopy, as well as pulse field gradient (PFG) technique and magnetic resonance imaging (MRI) can provide detailed information about molecular organization, specific interactions and internal mobility of constituents [5,6].

1.1. Hydrogels and organogels: definition and history

We begin with definitions of a few terms used throughout this review [7].