NMR spectroscopy is a very powerful technique for the investigation ofmolecular structure, conformation and dynamics. This thesis describes a detailed study of NMR diffusion processes in several systems and also includes the construction of novel 2D and 3D pulse sequences in the field of diffusion NMR. The main objective was to carry out detailed experiments to measure accurate self-diffusion coefficients of solution systems ranging in size from molecules to micelles. Diffusion-ordered spectroscopy (DOSY) is based on the pulse-field gradient spin-echo NMR sequence and can be used to separate the individual NMR spectra of a molecules in a mixture according to their diffusion coefficients. The strength of DOSY is that it can be used as a non-invasive method to obtain both physical and chemical information. The 2D DOSY experiment has limited potential to differentiate between severely overlapped signals in a mixture, which can lead to inaccurate estimates of diffusion coefficients. Several 3D DOSY experiments have been designed that achieve resolution of overlaps by concatenating a diffusion pulse sequence with common 2D pulse sequences. We have designed 3D BEST-DOSY, 3D COMPACT-IDOSY and 3D MQ-DOSY type of pulse sequences to circumvent the overlap problem in several molecular mixtures. Chapter 1 This chapter deals with fundamentals of NMR spectroscopy, fundamentals of diffusion, a brief description of the pulsed-field gradient NMR and application of PFG NMR, the basic spin echo experiment, 2D DOSY and its applications. Chapter 2 This chapter describes a pulse sequence we developed called 3D BEST-DOSY. This method focuses on exploiting the sensitivity-enhanced BEST-HMQC technique to achieve resolution of overlaps in a novel 3D heteronuclear 13C-1H diffusion ordered experiment with good sensitivity and a substantial reduction in experiment time. This pulse sequence has been applied on a mixture of molecules with similar diffusion coefficients, a mixture of amino acids and a mixture of commercial gasoline. Chapter 3 This chapter describes a new 3D I-DOSY type of pulse sequence we developed called COMPACT-IDOSY which includes a diffusion encoding sequence within a sensitivity enhanced HMBC-type pulse sequence. The direct incorporation of diffusionencoding into the heteronuclear coherence transfer sequence gives a substantial time and sensitivity advantage over standard 3D DOSY methods. The scheme has been experimentally demonstrated on a rutin trihydrate and quercetin dihydrate mixture, wherein the diffusion coefficients of the two molecular species are very similar. Chapter 4 This chapter deals with the incorporation of a novelmultiple-quantum/single-quantum correlation experiment into a DOSY sequence which we call the MQ-DOSY pulse sequence. This pulse sequence has been applied on small molecules and achieves good resolution of severely overlapped multiplets, leading to the accurate estimation of diffusion coefficients. Chapter 5 This chapter shows that drugs that have little or no difference in diffusion coefficients in simple solution may readily be resolved in DOSY experiments in solutions containing micelles. Two fluorinated drugs prulifloxacin and pazufloxacin were chosen as a test case. Their diffusion properties and T1 and T2 relaxation properties were investigated in DMSO-D6 solutions, and in solutions containing micelles. Chapter 6 This chapter deals with the extraction of diffusion information using PFGNMR of two anti-oxidants: alpha-tocopherol and curcumin, diffusing inside DPPC lipid bilayer. The relevance of diffusion NMR study of lipid-soluble antioxidants in lipid bilayer membrane models and micellar structures is discussed. Chapter 7 A brief outline of the main results of the thesis are summarized and some prospects for future extensions of the work are described in this chapter. Appendix A brief tutorial on how to install and calibrate the Diff30 diffusion probe as well protocols for diffusion data processing using the Bruker Topspin softare as well as the DOSY Toolbox software package, are given in the Appendix.