its distribution: the treatment with an acrylic polymer gives a good fit ($S \approx 0.2$ mm/s$^{1/2}$), while the treatment with a silicon-based polymer does not.

doi:10.1016/j.mri.2007.01.043

Multiple correlation function approach to study the restricted diffusion under arbitrary magnetic field

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A long-standing problem of restricted diffusion under arbitrary magnetic field is reformulated in terms of multiple correlation functions of the reflected Brownian motion [1]. For a typical spin echo experiment, the phase $\ddot{u}$ accumulated by a diffusing spin is

$$\varphi = \gamma \int_0^T dt f(t) B(X_t)$$

where γ is the nuclear gyromagnetic ratio, T is the echo time, $f(t)$ and $B(\mathbf{r})$ represent the time dependence and spatial inhomogeneities of the magnetic field, and X_t is the Brownian motion with normal reflections on the boundary of the confining domain Ω. The macroscopic NMR signal is obtained by averaging the individual magnetization $\exp[i]$ over a large number of diffusing spins, or expectation $E = \mathbf{E}\{\exp[i\varphi]\}$.
A series expansion of the exponential function relates the signal to the moments of the random phase φ

$$\boldsymbol{E}\{\varphi^n/n!\} = \gamma^n \langle \boldsymbol{E}\{B(X_{t1}) \ldots B(X_{tn})\} \rangle_n$$

where the f-weighted time average $\langle h(t_1,\ldots t_n)\rangle_n$ of any function h is defined as

$$\langle h(t_1, \ldots, t_n)\rangle_n = \int_0^T dt_1 f(t_1) \int_{t1}^T dt_2 f(t_2) \ldots \int_{tn-1}^T dt_n f(t_n) h(t_1 \ldots t_n)$$

The multiple correlation function $\mathbf{E}\{B(X_{t1}) \ldots B(X_{tn})\}$ of the reflected Brownian motion X_t is found to be equal to the first diagonal element of the matrix product

$$\boldsymbol{B} exp[-D(t_2 - t_1)\boldsymbol{\Lambda}]\boldsymbol{B} exp[-D(t_3 - t_2)\boldsymbol{\Lambda}]\boldsymbol{B} \ldots exp \times [-D(t_n - t_{n-1})\boldsymbol{\Lambda}]\boldsymbol{B} \qquad (*)$$

where D is the free diffusion coefficient, and two matrices $\mathbf{B}$ and $\boldsymbol{\Lambda}$ are defined through the eigenfunctions $u_m(\mathbf{r})$ and eigenvalues λ_m of the Laplace operator with Neumann (or reflecting) boundary condition:

$$\boldsymbol{B}_{m,m'} = \int_\Omega d\mathbf{r} \mathbf{u}_m^*(\mathbf{r}) \mathbf{B}(\mathbf{r}) \mathbf{u}_{m'}(\mathbf{r}) \quad \boldsymbol{\Lambda}_{m,m'} = \boldsymbol{\delta}_{m,m'} \boldsymbol{\Lambda}_m$$

An increase of the eigenvalues with m ensures a rapid convergence of the relation (*) and enables one to truncate the matrices $\mathbf{B}$ and $\boldsymbol{\Lambda}$ for numerical computation. Note that the mixed (or relaxing) boundary condition can be easily implemented [1].
By this description, the original physical problem of finding the macroscopic signal for arbitrary magnetic field is reduced to the analysis of the matrices $\mathbf{B}$ and $\boldsymbol{\Lambda}$. They can be found explicitly for basic domains like slab, cylinder or sphere, or computed numerically in the case of porous media. From the numerical point of view, this method appears as an extension of the multiple propagator or step-wise gradient approaches [2,3] for any spatial distribution of the magnetic field. But the crucial point is that the present technique allows one to study the restricted diffusion theoretically, whatever the confining geometry. The signal attenuation in the slow diffusion and motional narrowing regimes [4–6] has been retrieved in a more general form [1]. For instance, the results by Mitra et al. have been extended to any geometry and arbitrary temporal profile of the magnetic field:

$$D_{app} = D\left(1 - \mu_2 (DT/L^2)^{1/2} \langle (t_2 - t_1)^{3/2} \rangle_2 / \langle (t_2 - t_1) \rangle_2\right)$$

where μ_2 is a structure-dependent coefficient, L is the characteristic dimension of the confining domain, and the last factor accounts for the temporal profile. The determination of the surface-to-volume ratio in realistic porous media and possible ambiguities is discussed.
As a theoretical tool, this *multiple correlation function approach* allows one to examine, for example, the Gaussian phase and local gradient approximations. For practical applications, it opens a number of promising perspectives: careful accounting for nonlinear magnetic fields due to hardware imperfections or susceptibility effects; design and optimization of the magnetic field temporal and spatial profiles to enhance specific properties of diffusive motion.

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doi:10.1016/j.mri.2007.01.044

MR studies of the drying of surfactant granules

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The use of spray drying is well established in industry, particularly in the food and detergents industry, and is suited for the continuous production of dry solids in powder, granulate or agglomerate form. This work investigates the drying properties of an initially water-rich detergent mixture which features linear alkylbenzene sulphonate (LAS) as the active surfactant. The drying behaviour of the detergent mixture is important to the operation and optimisation of the spray drying process and to the resulting dried granule properties. Our work is concentrated on developing a more thorough understanding of this drying behaviour of the detergent mixture to aid both downstream processing and future applications. Whilst analogous information can be gained from other methods (e.g., rheology studies, mass spectrometry and infrared spectroscopy), the chemical specificity of nuclear magnetic resonance (MR) permits the roles of various components within the mixture to be studied simultaneously.
Various MR techniques have been used to study and hence map the spatially resolved drying and resultant phase changes that occur during drying. This has been performed on both vials filled with the detergent mixture (hence pseudo 1-D drying) and on in situ suspended droplets of the mixture drying in a heated air stream, hence simulating typical conditions with a spray drying process. Drying rates have been examined using both 1-D spin-echo and 1-D single point imaging techniques. Gravimetric measurements fall within these two measurement techniques; as expected, due to relaxation signal loss and ^{1}H signal from the LAS and other solid-like constituents. It has been determined that the drying rate is not dependent on external mass transfer and that it increases with temperature. Changes in the constituent phases of the detergent mixture have been monitored through analysis of T_1, T_2 and diffusion coefficient