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Appendix E1

The typical multicomponent spin-echo T2-mapping technique requires acquisition of images at varying echo time, and the image data are fit using a multiexponential decay model. In comparison, the mcDESPOT method fits observed spoiled gradient-recalled echo and balanced steady-state free precession signals at varying flip angle. The observed spoiled gradient-recalled echo and balanced steady-state free precession signals can be modeled by using a two-pool water exchanging system at steady-state condition as described below.

Consider a composite spin system including two exchanging water pools (fast (F) and slow (S) relaxing water pools) described in the mcDESPOT method. On the basis of the Bloch-McConnell equation, the spin evolution of magnetization can be expressed as follows:

$$\frac{dM}{dt} = AM + C \,,$$

where vector $M = \begin{bmatrix} M_{x,F} & M_{y,F} & M_{y,S} & M_{z,F} & M_{z,S} \end{bmatrix}^{T}$ contains components of the transverse (x, y) and longitudinal (z) magnetizations of each pool, and vector $C = \rho \begin{bmatrix} 0 & 0 & 0 & R_{1,F}f_F & R_{1,S}f_S \end{bmatrix}^{T}$. Here, 'T' denotes the transpose operation and ρ is the proton density term, f_F and f_S are fractional magnetizations for fast and slow water proton pools normalized by total water magnetization, respectively. $R_I = 1/T_1$ is longitudinal relaxation rate. *d* is the notation symbol for differentiation , *t* is time. All relaxation and exchange terms are included into a matrix, *A*, defined as follows:

$$\begin{split} A = & \begin{pmatrix} A_{\rm l} & 0 \\ 0 & A_{\rm 2} \end{pmatrix} \\ A_{\rm l} = & \begin{pmatrix} -R_{2,{\rm F}} - k_{{\rm FS}} & k_{{\rm SF}} & \Delta \omega & 0 \\ k_{{\rm FS}} & -R_{2,{\rm S}} - k_{{\rm SF}} & 0 & \Delta \omega \\ -\Delta \omega & 0 & -R_{2,{\rm F}} - k_{{\rm FS}} & k_{{\rm SF}} \\ 0 & -\Delta \omega & k_{{\rm FS}} & -R_{2,{\rm S}} - k_{{\rm SF}} \end{pmatrix} \\ A_{\rm 2} = & \begin{pmatrix} -R_{{\rm I},{\rm F}} - k_{{\rm FS}} & k_{{\rm SF}} \\ k_{{\rm FS}} & -R_{{\rm I},{\rm S}} - k_{{\rm SF}} \end{pmatrix}, \end{split}$$

where k_{SF} , k_{FS} are rates of (diffusion-driven) magnetization exchange between S and F pools and vice versa, respectively, and $\Delta \omega$ is the off-resonance frequency caused by B₀ field inhomogeneity. $R_2 = 1/T_2$ is the transverse relaxation rate. At chemical equilibrium, the fractional magnetization and exchange rates are related by:

$$k_{\rm FB}f_{\rm F} = k_{\rm SF}f_{\rm S}$$

For the balanced steady-state free precession signals (bSSFP) signal, an analytical solution of the above equation can be derived for the steady-state magnetization, $M_{\rm bSSFP}^{\rm SS}$:

$$M_{\rm bSSFP}^{\rm SS} = \left[I - e^{\rm A \cdot TR} R_{\alpha}\right]^{-1} \left(e^{\rm A \cdot TR} - I\right) A^{-1}C$$

where $M_{bSSFP}^{SS} = \begin{bmatrix} M_{x,F}^{SS} & M_{x,S}^{SS} & M_{y,F}^{SS} & M_{y,F}^{SS} & M_{z,F}^{SS} \end{bmatrix}^{T}$, *I* is the 6 × 6 identity matrix, TR is the sequence repetition time and matrix R_{α} is a rotation matrix with the prescribed flip angle α , expanded as follows:

$$R_{\alpha} = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & \cos \alpha & 0 & \sin \alpha & 0 \\ 0 & 0 & 0 & \cos \alpha & 0 & \sin \alpha \\ 0 & 0 & -\sin \alpha & 0 & \cos \alpha & 0 \\ 0 & 0 & 0 & -\sin \alpha & 0 & \cos \alpha \end{pmatrix}$$

The observed bSSFP signal, M_{bSSFP}^{obs} , is calculated as the magnitude of the complex summation of the transverse magnetizations of water components F and S:

$$M_{\text{bSSFP}}^{\text{obs}} = \left| \left(M_{\text{x,F}}^{\text{SS}} + i M_{\text{y,F}}^{\text{SS}} \right) + \left(M_{\text{x,S}}^{\text{SS}} + i M_{\text{y,S}}^{\text{SS}} \right) \right|$$

Spoiled gradient-recalled echo (SPGR) signal can be derived in a similar fashion. An analytical solution to the equation for the steady-state signal at SPGR sequence, $M_{\text{SPGR}}^{\text{SS}}$, can be expressed in the matrix form yielding the steady-state magnetization:

$$M_{\rm SPGR}^{\rm SS} = \rho \Big[I - e^{A_2 \cdot TR} \cos \alpha \Big]^{-1} \Big(I - e^{A_2 \cdot TR} \Big) \Big[\frac{f_{\rm F}}{f_{\rm S}} \Big]$$

where $M_{\text{SPGR}}^{\text{SS}} = \begin{bmatrix} M_{z,F}^{\text{SS}} & M_{z,S}^{\text{SS}} \end{bmatrix}^{\text{T}}$, and *I* is now the 2 × 2 identity matrix. The observed SPGR signal, $M_{\text{SPGR}}^{\text{obs}}$, can be calculated as follows:

$$M_{\rm SPGR}^{\rm obs} = \sin \alpha \left(M_{\rm z,F}^{\rm SS} + M_{\rm z,S}^{\rm SS} \right)$$