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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 $M = \begin{bmatrix} M_{x,F} & M_{x,S} & M_{y,F} & M_{y,S} & M_{z,F} & M_{z,S} \end{bmatrix}^T$ dt
 $M = \begin{bmatrix} M_{x,F} & M_{x,S} & M_{y,F} & M_{y,S} & M_{z,F} & M_{z,S} \end{bmatrix}^T$ conta contains components of the transverse (x, y) and longitudinal (z) magnetizations of each pool, and vector $P_{\text{1,F}} f_{\text{F}}$ $R_{\text{1,S}} f_{\text{S}}$]^T transverse (x, y) and longitudinal (z) magnetizations of each pool, and vector
 $C = \rho \begin{bmatrix} 0 & 0 & 0 & 0 & R_{1,F} f_F & R_{1,S} f_S \end{bmatrix}^T$. 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:

$$
A = \begin{pmatrix} A_1 & 0 \\ 0 & A_2 \end{pmatrix}
$$

\n
$$
A_1 = \begin{pmatrix} -R_{2,F} - k_{FS} & k_{SF} & \Delta\omega & 0 \\ k_{FS} & -R_{2,S} - k_{SF} & 0 & \Delta\omega \\ -\Delta\omega & 0 & -R_{2,F} - k_{FS} & k_{SF} \\ 0 & -\Delta\omega & k_{FS} & -R_{2,S} - k_{SF} \end{pmatrix}
$$

\n
$$
A_2 = \begin{pmatrix} -R_{1,F} - k_{FS} & k_{SF} \\ k_{FS} & -R_{1,S} - k_{SF} \end{pmatrix},
$$

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, $M_{\text{bSSFP}}^{\text{SS}}$:

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

where $M_{\text{bssFP}}^{\text{SS}} = \begin{bmatrix} M_{\text{v}}^{\text{SS}} & M_{\text{v}}^{\text{SS}} & M_{\text{v}}^{\text{SS}} & M_{\text{v}}^{\text{SS}} & M_{\text{z}}^{\text{SS}} & M_{\text{z}}^{\text{SS}} \end{bmatrix}$ $\mathcal{L}_{\text{bSSFP}}^{\text{SS}} = \begin{bmatrix} M_{\text{x,F}}^{\text{SS}} & M_{\text{x,S}}^{\text{SS}} & M_{\text{y,F}}^{\text{SS}} & M_{\text{y,S}}^{\text{SS}} & M_{\text{z,F}}^{\text{SS}} & M_{\text{z,S}}^{\text{SS}} \end{bmatrix}^{\text{T}}$ $M_{\text{bSSFP}} = \begin{bmatrix} I - e & K_{\alpha} \end{bmatrix} \begin{bmatrix} e & -I \end{bmatrix} A C$
 $M_{\text{bSSFP}}^{\text{SS}} = \begin{bmatrix} M_{\text{x,F}}^{\text{SS}} & M_{\text{x,S}}^{\text{SS}} & M_{\text{y,F}}^{\text{SS}} & M_{\text{y,S}}^{\text{SS}} & M_{\text{z,F}}^{\text{SS}} & M_{\text{z,S}}^{\text{SS}} \end{bmatrix}^{\text{T}}$, *I* is the , *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:

llows:

\n
$$
R_{\alpha} = \begin{pmatrix}\n1 & 0 & 0 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 & 0 & 0 \\
0 & 0 & \cos \alpha & 0 & \sin \alpha & 0 \\
0 & 0 & 0 & \cos \alpha & 0 & \sin \alpha \\
0 & 0 & -\sin \alpha & 0 & \cos \alpha & 0 \\
0 & 0 & 0 & -\sin \alpha & 0 & \cos \alpha\n\end{pmatrix}
$$

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

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_{SPGR}^{SS} , can be expressed in the matrix form yielding the steady-state magnetization:):
 $\left\lceil f_{\textrm{F}}\right\rceil$

$$
M_{\text{SPGR}}^{\text{SS}} = \rho \Big[I - e^{A_2 \cdot \text{TR}} \cos \alpha \Big]^{-1} \Big(I - e^{A_2 \cdot \text{TR}} \Big) \Big[\frac{f_F}{f_S} \Big]
$$

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

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