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An Advanced Calibration Strategy for *in-situ* Quantitative Monitoring of Phase Transition Processes in Suspensions Using FT-Raman Spectroscopy

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MATLAB code for the estimation of multiplicative parameters q_k by OPLEC

% [**q**] = OPLEC(**Z**, **y**, CompNumb);
% This is an m-file to find the multiplicative scattering parameter vector **q** for calibration samples;
% **Z** contains $\mathbf{I}_k$ in its rows; $\mathbf{I}_k$ $(k = 1,2,\cdots,K)$ are the Raman spectra of calibration samples.
% **y** is the concentration vector of the target chemical component in calibration samples;
% CompNumb is the number of chemical components in mixture samples;
% **q** is a vector containing the multiplicative scattering parameters q_k for calibration samples;

```
function [q]=OPLEC(Z,y,CompNumb);

[m,n]=size(Z);
Y=[ones(m,1),y];
q=zeros(m,m);

for i=1:m

    OriginalPositionIndex=(1:m)';
    Z1=Z(i,:);
    PositionIndex=i;

    for j=1:(CompNumb-1)
        NormVect=sqrt(sum(Z.^2,2));
        ResMatrix=diag(1./NormVect)*Z*(eye(n,n)-pinv(Z1)*Z1);
        ResVect=diag(ResMatrix*ResMatrix');
        [MaxRes,MaxPosition]=max(ResVect);
        Z1=[Z1;Z(MaxPosition,:)];
        PositionIndex=[PositionIndex,MaxPosition];
    end

    OriginalPositionIndex(PositionIndex)=[];

    ZZ=Z;
    ZZ(PositionIndex,:)=[];
    Z2=ZZ;
```

```
    Y11=Y(PositionIndex,1);
    Y21=Y(PositionIndex,2);
    YY=Y;
    YY(PositionIndex,:)=[];
    Y12=YY(:,1);
    Y22=YY(:,2);

    Regv=Z2*pinv(Z1);
    P1=diag(Y22)*Regv*diag(Y11);
    P2=Regv*diag(Y21);
    P11=P1(:,1);
    P12=P1(:,2:CompNumb);
    P21=P2(:,1);
     P22=P2(:,2:CompNumb);

    q0=lsqnonneg(P12-P22,P21-P11);
    q1=[1;q0];

    Q1=Regv*diag(Y11);
    Q2=Regv*diag(Y21);
    q2=diag(1./(Y12+Y22))*(Q1+Q2)*q1;

    q(PositionIndex,i)=q1;
    q(OriginalPositionIndex,i)=q2;

end

q=mean(q,2);
```

% After obtaining the model parameter vector **q** for calibration samples, two calibration models are built by standard PLS toolbox. One is between the concentration vector ($\mathbf{y}$) of the target chemical component and the spectral data **Z**, the other is between $diag(\mathbf{y})\mathbf{q}$ and **Z**. The multiplicative effect on the test sample can then be corrected through dividing the prediction of the second calibration model by the prediction of the first calibration model.