Supplementary Material (ESI) for Journal of Materials Chemistry

Supporting Information for:

In Situ Neutron Diffraction Study of the High-Temperature Redox Chemistry of $Ln_{3-x}Sr_{1+x}CrNiO_{8-\delta}$ (*Ln* = La, Nd) under Hydrogen

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Figure S1. Thermogravimetric analysis (heating rate of 2 °C min^{-1}) under 5 % H_2 in N_2 for $Nd_{2.25}Sr_{1..75}CrNiO_{8-\delta}$

Figure S2. XRD pattern fit at room temperature of $La_2Sr_2CrNiO_{8-\delta}$; (a) as-prepared, (b) after hydrogen reduction (in 5% H_2 at 700 °C, 8h)

Figure S3. XRD pattern fit at room temperature of $Nd_2Sr_2CrNiO_{8-\delta}$; (a) as-prepared, (b) after hydrogen reduction (in 5% H_2 at 700 °C, 8h)

Figure S4. XRD pattern fit at room temperature of $Nd_{2.25}Sr_{1.75}CrNiO_{8-\delta}$; (a) as-prepared, (b) after hydrogen reduction (in 5% H_2 at 700 °C 8h)

Figure S5. Neutron Powder Diffraction pattern fit of $Nd_2Sr_2CrNiO_{8-\delta}$; (a) at 20 °C before hydrogen reduction, (b) at 710 °C after reduction, (c) at 90 °C after reduction and cooling

Figure S6. Neutron Powder Diffraction pattern fit of $Nd_{2.25}Sr_{1.75}CrNiO_{8-\delta}$; (a) at 20 °C before hydrogen reduction, (b) at 450 °C after reduction, (c) at 80 °C after reduction

Figure S7. Occupancy factor of oxygen sites in (a) $Nd_2Sr_2CrNiO_{8-\delta}$ and (b) $Nd_{2.25}Sr_{1.75}CrNiO_{8-\delta}$ as a function of temperature under hydrogen flow. Green and black squares represent the occupancy of O1 (axial) and O2 (equatorial) sites during heating; blue and red triangles represent O1 and O2 on cooling. The blue line corresponds to the Ni (II) composition

Figure S8. Temperature dependence of the unit cell parameters of (a) $Nd_2Sr_2CrNiO_{8-\delta}$ and (b) $Nd_{2.25}Sr_{1.75}CrNiO_{8-\delta}$ under hydrogen flow. Squares (triangles) represent data collected on heating (cooling).

Figure S9. (a) Occupancy factor of oxygen sites in $Nd_2Sr_2CrNiO_{8-\delta}$ as a function of temperature under oxidizing conditions after reduction. Green and black squares represent the occupancy of O1 (axial) and O2 (equatorial) sites during heating; blue and red triangles represent O1 and O2 during cooling;

(b) temperature dependence of the unit cell parameters of $Nd_2Sr_2CrNiO_{8-\delta}$ under oxidizing conditions. Squares (triangles) represent data collected on heating (cooling)

Figure S10 Rietveld refinements of the X ray diffraction pattern of $Nd_2Sr_2CrNiO_{7.38}$ between 2θ = 76.6° and 81.1° (a) with a single profile for all peaks, (b) with anisotropic peak-broadening taken into account.

Figure S11. Temperature dependence of the (a) Cr/Ni-O1, (b) Nd/Sr-O1 (along the *c*-axis) and (c) Nd/Sr-O2 bond lengths under hydrogen flow, in $Nd_2Sr_2CrNiO_{8-\delta}$. Squares (triangles) represent data collected on heating (cooling)

Figure S12. Temperature dependence of the (a) Cr/Ni-O1, (b) Nd/Sr-O1 (along the *c*-axis) and (c) Nd/Sr-O2 bond lengths under hydrogen flow, in $Nd_{2.25}Sr_{1.75}CrNiO_{8-\delta}$. Squares (triangles) represent data collected on heating (cooling)

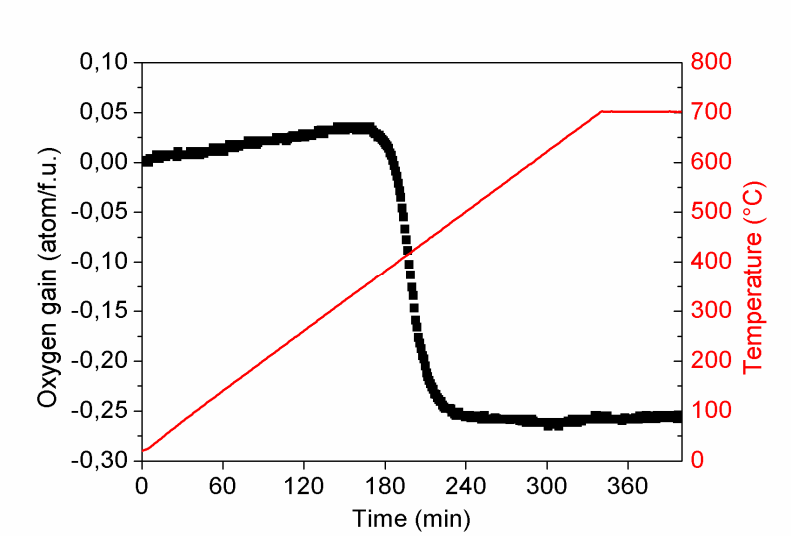
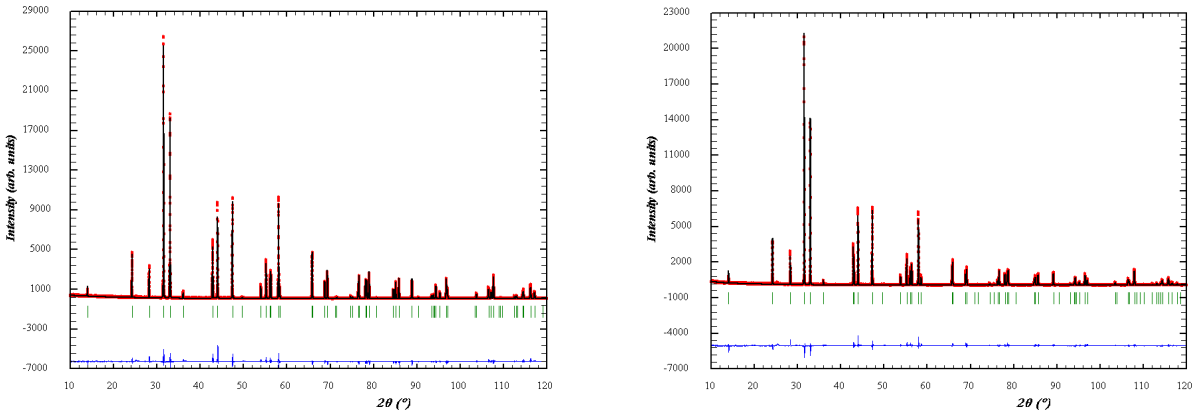
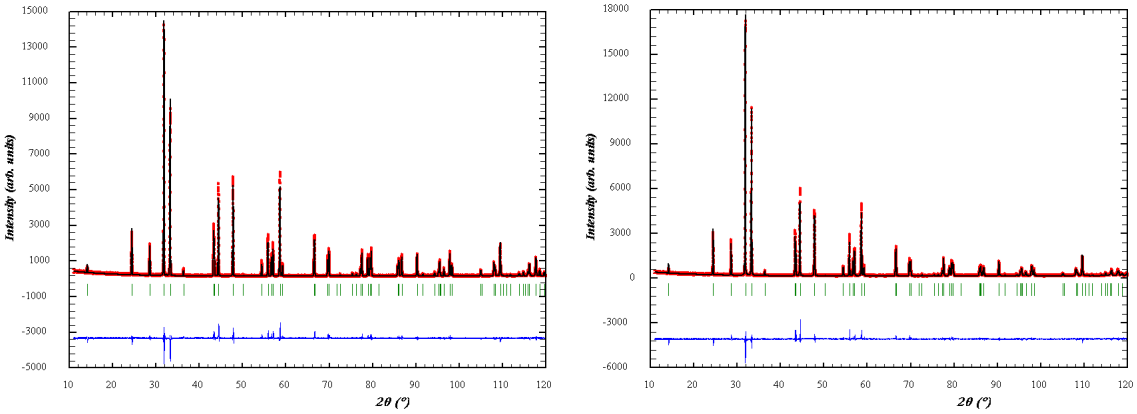


Figure S1



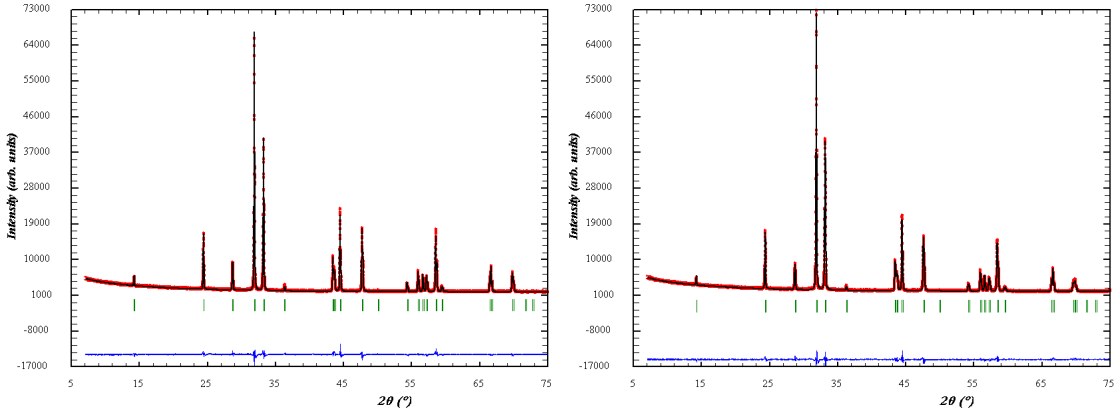
(a) **(b)**

Figure S2



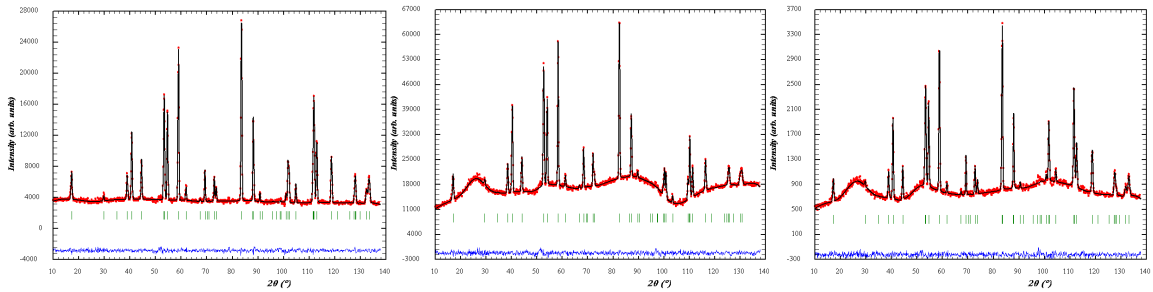
(a) **(b)**

Figure S3



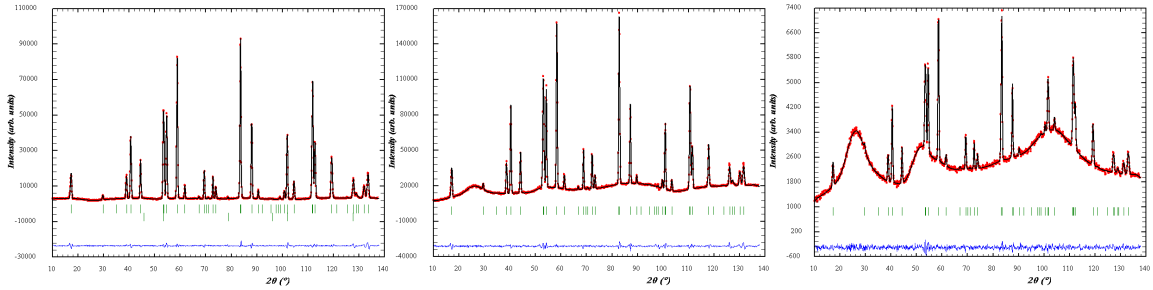
(a) **(b)**

Figure S4



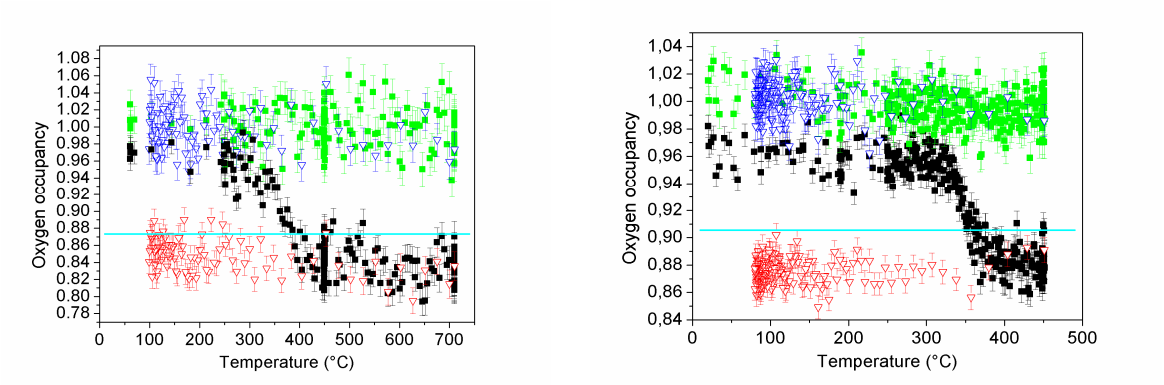
(a) **(b)** **(c)**

Figure S5



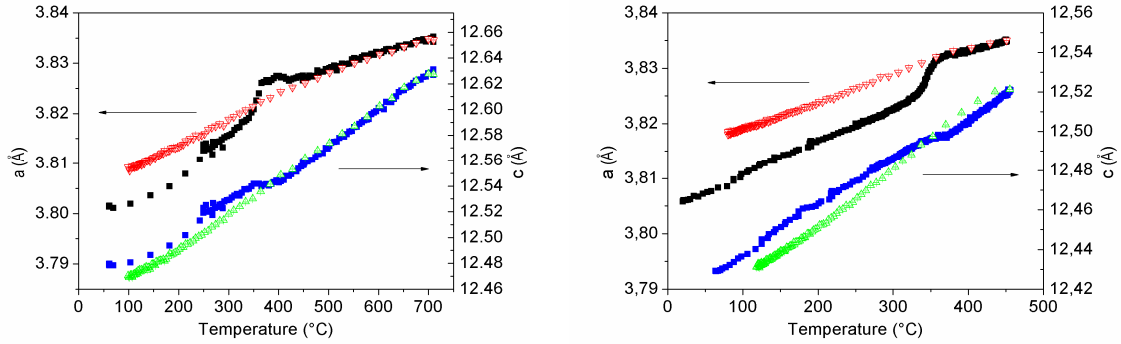
(a) **(b)** **(c)**

Figure S6



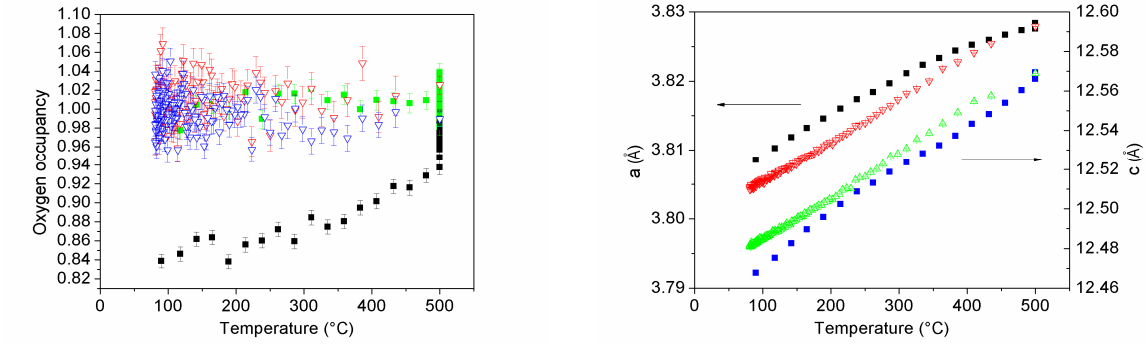
(a) **(b)**

Figure S7



(a) **(b)**

Figure S8



(a) **(b)**

Figure S9

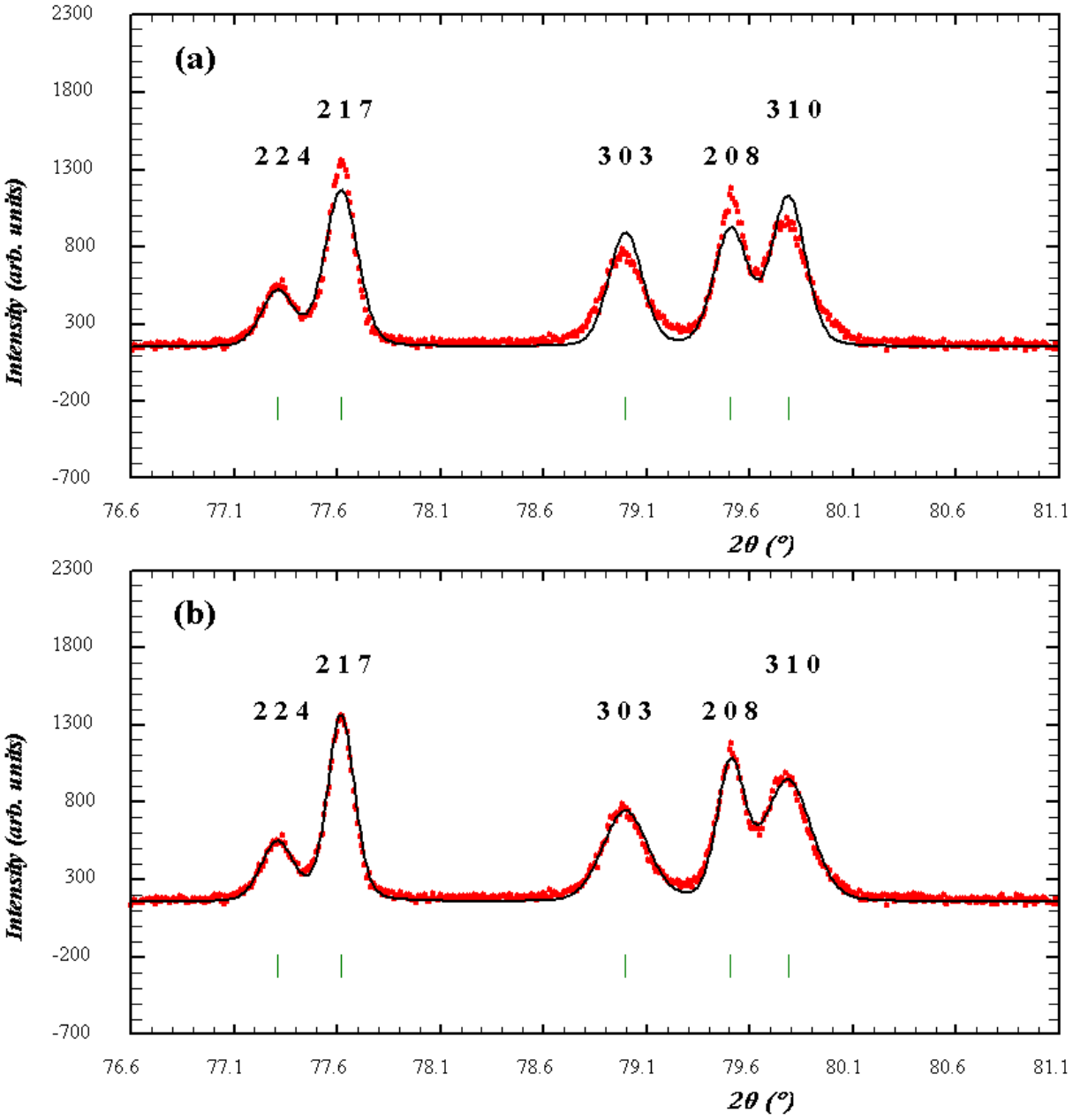
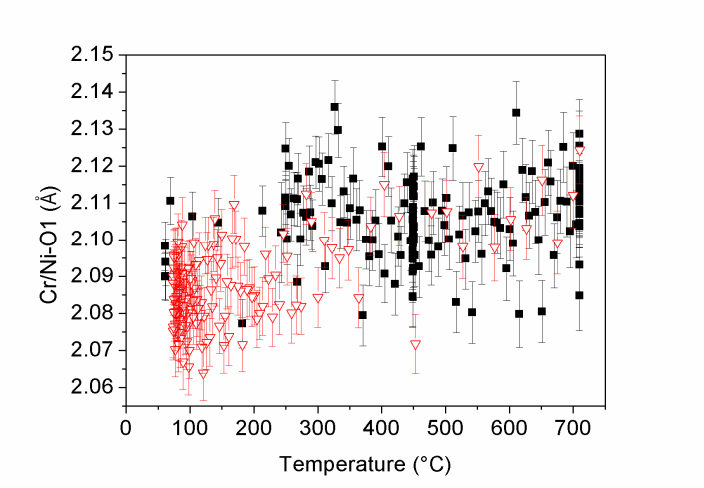
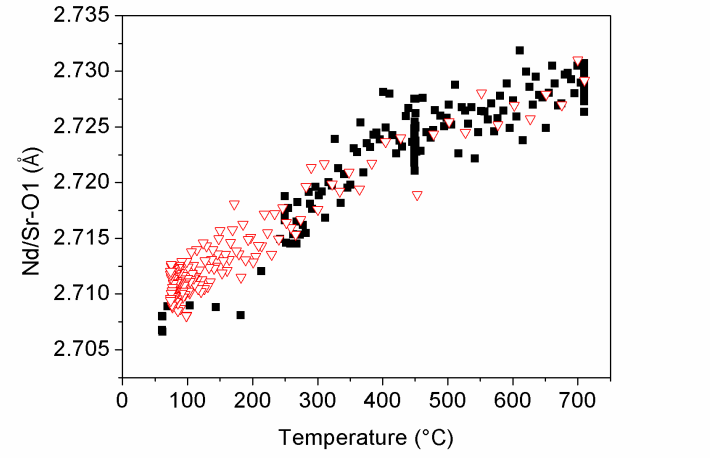


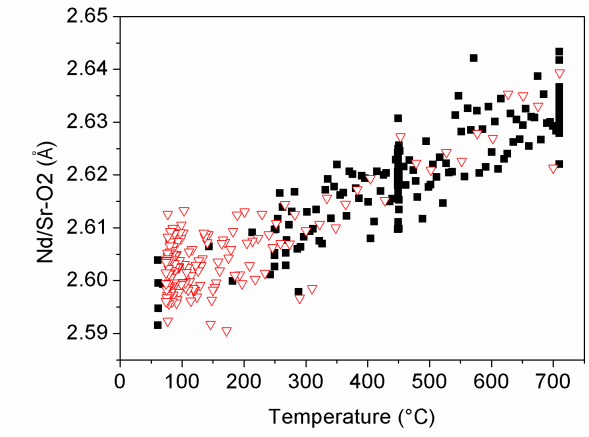
Figure S10



(a)

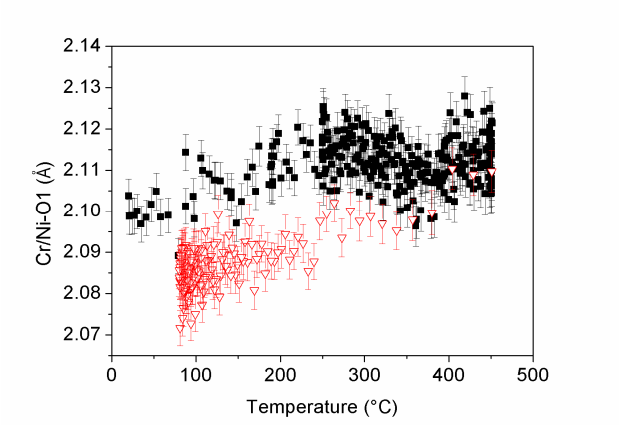


(b)

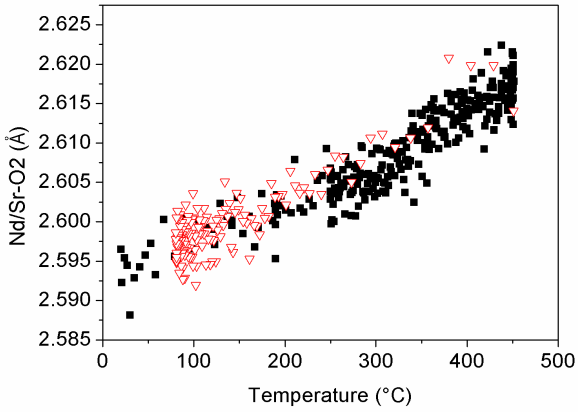


(c)

Figure S11



(a)



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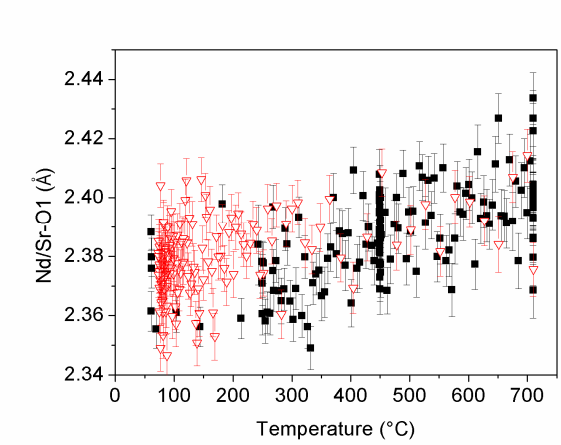

(c)

Figure S12