Calculation of the orientation relationships of directionally solidified eutectic ceramics by a modified Coincidence of Reciprocal Lattice Points model (CRLP)

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Abstract

The Coincidence of Reciprocal Lattice Points (CRLP) method was used to predict, according to geometric considerations, the most favorable orientation relationships between the component phases in a family of directionally solidified eutectic ceramics (NiO-YSZ, CoO-YSZ, NiO-CeO_2, NiO-GDC, CoO-CeO_2 and CoO-GDC) grown by the laser-floating-zone method. The orientation relationships predicted by the CRLP model are consistent with those experimentally found in a previous work by means of Electron Backscatter Diffraction (EBSD). In this paper we also present a modification to the model with the aim of taking into account that the most stable ceramic-ceramic interfaces are usually formed between atomic planes with low-Miller indices, due to their higher atomic density and bigger interplanar spacing. Thus, we introduce in the calculation of the overall coincidence volume a weighting factor which is a function of the interplanar spacing. This modified CRLP method has been applied to the aforementioned eutectic ceramics, and the results are presented and discussed in comparison with the traditional CRLP results and the experimental findings.

1. Introduction

Properties of most composite materials are strongly influenced by the interface and orientation relationships between phases. Highly textured materials, such as Directionally Solidified Eutectic Ceramics (DSEC), show significant differences in their properties when compared to materials with the same composition but different fabrication methods.[1] DSEC's are self-organized materials with coherent and homogeneous microstructures. The orientation relationship (OR) and interfacial plane stem from the minimization of the interfacial energy, whereas the size of the phases may be tailored, within certain limits, by selecting the solidification rate according to the Hunt-Jackson law.[2] In this way we can obtain materials with very interesting and unusual properties, for instance the nanofibrillar Al_2O_3-YAG-ZrO_2 ternary eutectic which presents outstanding mechanical properties, 4.6 GPa of flexure strength and 4.7 $MPa \cdot m^{1/2}$ of fracture toughness, because of its unique microstructure.[3]

Binary DSEC's made up of a transition metal oxide (NiO or CoO) and an ionic conductor (YSZ: yttrium-stabilized zirconia, CeO_2 or GDC: gadolinium-doped ceria), display a highly textured self-organized lamellar microstructure with fixed ORs between phases. This type of microstructure results in a strong adhesion between the eutectic components. After reduction of the transition metal oxide, the cermets which are obtained are formed by alternate lamellae of an ionic conductor and a porous metal. The good adhesion that is obtained in DSE cermets induces a great stability of the metallic particles in the porous cermet against coarsening during long-term operation at high temperature. Besides, these cermets have easy gas flow, high electric conductivity as well as thermal, chemical and mechanical stability at high temperatures. Thus they have good properties to be used as anodes in Solid Oxide Fuel Cells (SOFC). [4-5] Understanding the microstructure and the nature of the interfaces of these materials,

before and after reduction, is essential to assess their suitability and performance in an SOFC.

The state-of-the-art anode in an SOFC is a porous cermet of Ni and YSZ. [6] One of the concerns with these anodes is its long-term stability, because Ni particles tend to agglomerate into larger particles during cell operation, degrading the cell functionality. This material, when obtained through reduction of DSEC NiO-YSZ, is more stable than when obtained via the traditional process of reduction of a sintered mixture of NiO and YSZ. [7] The main difference between these two materials is that NiO and YSZ phases resulting from directional solidification show a preferential orientation relationship [8] and well-defined interfaces, whereas interfaces and OR's in sintered NiO-YSZ are random. Similar behavior was observed when comparing traditional Co-GDC cermets vs Co-GDC cermets obtained from the CoO-GDC DSEC. [9]

Ceramic-ceramic and metal-ceramic orientation relationships and interfaces have been extensively studied for many systems, both from the experimental and the theoretical point of view. [10, 11] As for the theoretical studies on epitaxial interfaces, density functional theory (DFT) calculation is successfully being applied to an increasing number of materials and interfaces.[12,13] However, to apply the DFT calculation to DSECs the particular interface has to be known *a priori* and a large calculation time is also needed. Nevertheless, we would like to have a tool able to predict the actual orientation relationship occurring in the different DSEC's from minimum previous knowledge. For this purpose we will consider that the particular OR and interface between the component phases are selected by the minimization of the interfacial energy during the eutectic coupled growth. In such a context, a purely geometric model might be useful. To this aim, there have been numerous attempts to establish a universal model able to predict orientation relationships in heterophase interfaces from

geometrical considerations.[15] The most relevant ones are the Near Coincident Site Lattice (NCSL) model, the Edge-to-Edge Matching model and the Coincidence of Reciprocal Lattice Points (CRLP) model. They have proved useful to account for orientation relationships in some materials, but a universal model is far from having been achieved and will probably never be developed from purely geometrical calculations, since the component phases chemistry should also be considered.

The NCSL model is a modification to the Coincident Site lattice (CSL) model,[15] which was designed for grain boundaries, so that the same principles could be applied to interphase boundaries.[16] These models are based on the assumption that a grain boundary is a low-energy one when the adjoining crystal lattices are at a relative orientation such that there is a high density of coincident lattice sites if we superpose the lattices (or nearly-coincident lattice sites if we consider an interphase boundary). For these calculations to be applied, the interface planes must be known. Otherwise, every possible interface planes combination would have to be tested, thus dramatically increasing computational times.

The Edge-to-Edge Matching model was devised by Kelly and Zhang in 1998[17] as a way to account for orientation relationships and interface planes in partially coherent precipitates. It postulates that minimization of the interfacial energy takes place when close-packed, or nearly close-packed, rows of atoms from the two phases, with similar spacing between the atom rows, match at the interface, edge-to-edge. It has been successfully applied to fcc/bcc and simple hcp/bcc compounds.[18]

The CRLP model is based upon the hypothesis that most stable configurations correspond to relative orientations where the crystal lattices suffer minimum distortion across the boundary in terms of parallelism and interplanar spacing continuity. In other

words, those orientation relationships where there are many families of planes parallel to each other, and with similar interplanar spacings, are favoured, and therefore the phases will tend to orientate according to these orientation relationships.[19] The underlying geometrical criterion is quite similar to that of the NCSL model, but the CRLP model might easily be applied even when the interface planes are unknown. In fact, the CRLP model was developed to study orientation relationships between phases away from their interface[20] and it has been successfully applied to account for orientation relationships in numerous systems, such as AlN/SiC[21], Si/Al_2O_3, [20] Nb/$BaTiO_3$,[22] $BaTiO_3$,[23] MgO,[24] Au/TiO_2,[25] Ti/6H-SiC,[26] $LiNb_{0.5}Ta_{0.5}O_3$/sapphire,[27] Ni/YSZ,[28] $Ba_{0.6}Sr_{0.4}TiO_3$,[29] SiC/$MgAl_2O_4$/Al,[30] TiB/ γ-TiAl[31] and Al-Si/AlN.[32]

Other geometrical models less frequently used to predict orientation relationships are the lock-in model, the planar CSL model combined with the largest interplanar spacing, the **Δg** parallelism rules, the symmetry-dictated criterion and the invariant line criterion. The lock-in-model was first developed by Fecht and Gleiter[33] for heterophase boundaries between a metal surface and an ionic crystal. According to this model, a low-energy boundary occurs when a close-packed direction in the crystal lattice of one phase is parallel to a close-packed direction in the crystal lattice of the other phase. The metal crystal rotates about this common direction so that sets of low-index lattice planes are parallel in both phases. The close-packed rows of metal atoms are deemed to lie in the valleys between the close-packed rows of the ionic crystal. This model was successfully applied to account for orientation relationships in several ceramic-Au and ceramic-Cu systems (LiF-, MgO-, NaCl-, KCl-, mica-, Al2O3-Au and MgO-, Al_2O_3-Cu).

Wolf[34] developed the planar CSL model combined with the largest interplanar spacing criterion as a modification to the two-dimensional CSL model. This model postulates that not only a good in-plane coincidence between the sites in both lattices favors the appearance of a given orientation relationship (as a grain boundary or an interphase boundary), but also the planes with the largest interplanar spacing, or in other words, the planes with the highest atomic density, are more prone to form the boundary between phases. This thesis is inferred from Pauling's principle, which states that all interatomic interaction potentials increase sharply (almost exponentially) with decreasing separation between two atoms.[35] Thus, the boundary is expected to decrease its energy as the interplanar spacing increases.

The **Δg** parallelism rules, where **Δg** is a difference vector linking correlated reciprocal lattice vectors from two adjacent phases, were proposed by Zhang and Weatherly.[36] These rules help to identify local minima of the interfacial energy in systems in which well-defined facets were observed. It has been successfully applied to ceramic/metal and metal/metal interfaces. When compared to experimental observations, the authors found that most systems followed what they call Rule I, according to which, the major facets are parallel to a low index plane in one or both crystal lattices.

Cahn and Kalonji postulated the symmetry-dictated criterion.[37] According to it, the parallelism of symmetry axes common to both crystals may lead to minimum interfacial energy. The invariant line criterion, on the other hand, assesses the orientation relationships with an invariant line (a line of zero misfit) in parallel close-packed planes from both phases as the most favorable ones.[38] All parametric models are based on the hypothesis that the orientation relationships which optimize a certain parameter are the most favorable ones. In the case of the CRLP model, the parameter to maximize is the overlapping volume of the spheres located at reciprocal sites. Other parametric

models use different parameters, such as a function of the magnitude of the Burgers vector and the dislocation spacing.[39,40] Sutton and Balluffi[14] and Zhang and Weatherly[36] reviewed all the aforementioned geometrical models and their successful and unsuccessful applications to experimentally observed OR's more in depth. The parallelism between low-Miller-index planes appears repeatedly as an important geometric criterion throughout the models, either as parallelism between close-packed directions or parallelism between planes with the highest atomic density. Thus, in this paper we have tested a modification to the CRLP method in order to take into account the parallelism between low-Miller-index planes criterion, and we show the results of applying this model, with and without the modification, to a family of DSECs (NiO-YSZ, CoO-YSZ, NiO-GDC, NiO-CeO_2, CoO-GDC and CoO-CeO_2) prepared by the laser floating-zone technique. Growth directions, orientation relationships and interface planes for these materials were experimentally determined by means of Electron Back-Scattering Diffraction (EBSD) in a previous paper, in which the application of the CRLP method to the NiO-YSZ eutectic was advanced.[41]

2. Theory

The CRLP model is based on the idea that orientation relationships between phases with high continuity of the crystal lattice are favoured and thus, more likely to actually appear in multiphase materials. To measure the degree of "continuity" through the interphase boundary, we consider the parallelism between planes from both lattices and roughly similar interplanar spacing. To this aim, reciprocal lattice is very appropriate. Each family of the direct lattice planes (hkl) is associated with a point P in the reciprocal lattice as follows: $\boldsymbol{OP}_{hkl}=h\cdot\boldsymbol{a}^*+k\cdot\boldsymbol{b}^*+l\cdot\boldsymbol{c}^*$, where $\boldsymbol{a}^*$, $\boldsymbol{b}^*$ and $\boldsymbol{c}^*$ are the primitive vectors of the reciprocal lattice, the modulus of the $\boldsymbol{OP}_{hkl}$ vector being the inverse of the interplanar spacing.[42] The angle between direct lattice planes, (hkl) and

(h'k'l'), is the same as between their corresponding reciprocal lattice vectors, $\boldsymbol{OP_{hkl}}$ and $\boldsymbol{OP_{h'k'l'}}$. If we place imaginary spheres with radius r* at each point of the reciprocal lattices of both phases and superimpose both reciprocal lattices, we can calculate the overlapping volume of the spheres from the two phases. This overlapping volume is as a measurement of the degree of parallelism between direct lattice planes from both phases, as well as of the matching of their interplanar spacings. As a consequence, it is a measurement of the continuity of the crystal lattice through the boundary. The most favourable orientation relationships according to the CRLP model are those with the highest overlapping volume, since this means that there are several family planes in the direct lattice with similar interspacing parallel to each other, and therefore, there is little discontinuity at the interface formed by this orientation relationship.

It should be noted that the phases studied in this paper are cubic ones and they all show elastic anisotropy to a certain degree, being their Zener ratios 1.45 for NiO[i] and CoO,[ii] 0.47 for ZrO_2[iii] and 0.40 for CeO_2.[iv] However, elastic isotropy (Zener ratio = 1) is implicitly assumed in the CRLP model, given the fact that the same r* parameter is used for all the involved direct lattice planes. Consequently, the model validity for very anisotropic cubic or non-cubic systems should be evaluated considering how reasonable this assumption may be in each phase.

3. Calculations

The CRLP calculations were performed in Matlab code. For the calculation of the overlapping volume as a function of the OR, the transition metal oxide (NiO or CoO) lattice was set fixed and the ionic conductor phase (YSZ, CeO_2 or GDC) was rotated in steps of 5º using the Euler angles. We used the ZXZ´ convention for the Euler angle definition and reduced the Euler space to $\alpha \in (0º, 90º)$, $\beta \in (0º, 90º)$ and $\gamma \in (0º, 90º)$ due

to the cubic symmetry of the phases.[43] The crystallographic parameters of all the phases involved are shown in Table 1.

The total overlapping volume was estimated as the sum of the intersections between all spheres from crystal lattices A and B through the following expression:

$$V_{overlapping} = \sum_i V_{intersection}\left[Sphere_{hkl}^{Phase\ A}, Sphere_{h'k'l'}^{Phase\ B}\right] \cdot w_i, \qquad \text{Eq.(1)}$$

where $w_i = \left(d_{hkl} \cdot d_{h'k'l'}\right)^k$ is the weighting factor to enhance the importance of the parallelism between low-Miller-index planes, i.e. those with high atomic density and interplanar spacing, d_{hkl} . The value k=0 gives the results corresponding to the conventional unweighting CRLP method, whereas values of k=1, k=2 and k=3 were used to check the weighting version (see Table 2).

4. Results and discussion

In order to perform the calculations there are two parameters that need to be correctly adjusted: the size of the imaginary spheres located at the reciprocal lattice sites, r*, and the size of the reciprocal space considered, R*, i.e., the largest Miller index planes taken into account in the direct lattice. As Stemmer *et al.*[21] discussed, an increase in the value of R* or a decrease in the value of r*, results in an enhancement in the peak resolution. The range of values used for r* in the literature goes from 0.1·a*[21, 32] to 0.7·a*,[30] where a* is the inverse of the cell parameter, though most commonly values used for r* lie between 0.2·a* and 0.4·a*. CRLP calculations for CoO-YSZ with k=0 and R* = 10·a* were performed with r* values ranging from 0.01·a* to 0.4·a*. In Fig. 1, results for r*= 0.1·a*, 0.15·a*, 0.25·a* and 0.35·a* are shown. The horizontal axes correspond to the first two Euler angles, α and β, and in the vertical axis we represent the overlapping volume (in arbitrary units) for each pair of α and β values. Third Euler

angle, γ, has been set to 0º for the sake of clarity in the graphical display of results. It can be inferred that, for r* values below 0.2·a*, too much noise appears producing spurious local maxima. Therefore, a compromise must be set between peak resolution and acceptable level of noise. This process has been applied to the whole DSEC family studied and we concluded that r* values ranging from 0.2·a* to 0.25·a* are a good choice.

As for the size of the reciprocal space considered, R*, in spite of what Stemmer et al. said, most authors have restricted R* to 2 or 3 times a*. Only Montesa *et al.*[32] and Luo[30] used R* values as big as 10·a*. We also explored values of R* up to 10·a*. Results for some of these values can be seen in Fig.2. Two equally high peaks located at the centre of the graphs (α_1=45º, β_1=35.3º, γ_1=0º and α_2=45º, β_2=54.7º, γ_2=0º) can be observed. They correspond to the greatest overlapping volume for the calculations with R*>2a* and to the second maxima for the R*=2a* calculation. Taking into account the crystal symmetry of the phases, it can be shown that these two triplets of Euler angles are equivalent to each other and to 574 other Euler triplets. The set of symmetry equivalent Euler rotation matrices, G(α,β,γ), can be obtained by the following expression:

$$G(\alpha,\beta,\gamma)=O_i \cdot G'(\alpha',\beta',\gamma') \cdot O_j, \qquad \forall\, O_i, O_j \in R(m\bar{3}m) \qquad \text{Eq. (2)}$$

where O_i are the 24 proper rotations of the point group of the component phases ($m3\bar{3}m$). Thus, there are 24x24=576 symmetrically equivalent triplets of Euler angles that correspond to each individual OR. With respect to the aforementioned triplets, all other equivalent triplets do not have γ=0º and cannot be seen in Fig. 2. These triplets predicted by the CRLP method do correspond to the experimentally found majority orientation relationship (OR1 from now on) determined by EBSD in a previous work

[40]. This majority orientation relationship was found in all the DSEC's studied in that paper and is given by:

$(200)_{YSZ/CeO2/GDC}$ // $(111)_{NiO/CoO}$ (interfacial planes)

and $[001]_{YSZ/CeO2/GDC}$ // $[0\bar{1}1]_{NiO/CoO}$

In addition to OR1, a minority orientation relationship was also found in the EBSD experiments, the so-called cube-on-cube orientation relationship (OR2 from now on), where both cubic phases appear with their unit cells parallel to each other. Different EBSD experiments were carried out on transverse and longitudinal cross-sections of NiO-YSZ in order to determine the interface plan. Unfortunately, this could not be established with total accuracy, but it was discovered that the interface plane is approximately (0.185, 0.188, 0.965), which forms ~15º with the (001) plane. In Fig. 2 OR2 corresponds to the triplet α=0º, β=0º and γ=0º and equivalent points at each corner of the α-β plane. The relative height of the peaks representing OR2 depends largely on the value of R*. They are the absolute maxima for R=2a*, secondary maxima for R=6a* and R=10a* with very different relative importance and, even, the minimum for the R=8a* calculations. It is noteworthy that the CRLP results show a strong dependence on the size of the reciprocal space considered, R*. While the majority OR appears in all cases as the absolute maxima or, at least, a relevant relative one, other orientation relationships are represented by peaks that change, even from relative maximum to relative minimum, significantly as a function of R*. When R* is increased, the model results become inconsistent while the expected improvement of the peak resolution is doubtful. The reason why most authors have used such small reciprocal spaces (R* = 2·a* or 3·a*) and, as a consequence, overlooked possible parallelisms between high-Miller-index planes in the direct lattice, may be that such parallelisms are not usually

found in actual orientation relationships in heterophase boundaries. The conventional CRLP model attaches equal importance to parallelism between low-Miller-index planes and parallelism between high-Miller-index planes. By trimming the considered reciprocal space at a small radius R*, it is feasible that the appearance of spurious secondary relative is avoided. However, the criteria used to choose the reciprocal space size seem somewhat arbitrary.

Thus we explore in this paper a modification of the conventional CRLP method to enhance the relevance of the parallelisms between low-Miller-index planes by gradually diminishing the importance of coincidence between high-Miller-index reciprocal sites, without actually having to choose a cut-off interplanar spacing. As explained in section 3, the modification proposed consists of introducing a weighting factor, $w_i = \left(d_{hkl} \cdot d_{h'k'l'}\right)^k$, in the calculation of the overlapping volume (Eq. 1), which aims to increase the importance of parallelisms between close-packed low-Miller-index planes, since these are more likely to turn into actual orientation relationships. Fig. 3 shows the results of this modified method for CoO-YSZ eutectic with k=1 and R*=2·a*, 6·a*, 8·a* and 10·a*. It can be seen that the weighting procedure has a positive effect on the strong dependence of the CRLP model with the value of R*. With this there are no dramatic changes when we increase R*. Of course new coincidences appear and the relative peak height varies only slightly, but there are no orientation relationships which turn from relative minimum into maximum or vice versa, as sometimes happens with the conventional CRLP calculations. Therefore, these results are more consistent and we do not need to choose the smallest interplanar spacing to be considered, we can just set R* to a high enough value (15 or 20, for instance). Using a high enough R* value is also important when there is a significant difference between the lattice parameters of the phases, as we need to check that the reciprocal space considered includes enough points

from both phases for the results to be meaningful. Using the modified CRLP method we have been able to enlarge the reciprocal space in which we explore coincidences up to R*=20·a*. The results for k=1, 2 and 3 for CoO-YSZ are shown in Fig. 4, with r*=0.25·a*, R*=20·a* and γ=0. Calculations involving the full 3D Euler space to ensure that there are no other maxima with γ≠0º higher than those represented in Fig. 4 have also been performed. Similar results have been found for the other DSEC's studied: CoO-YSZ, NiO-CeO_2, NiO-GDC, CoO-CeO_2 and CoO-GDC. Fig. 5 displays the overlapping volume for them, with k=3, R*=20·a* and r*=0.25·a*. With these parameters, the most favourable orientation relationships according to the modified CRLP model do correspond to the experimentally found OR.

In some materials (NiO-GDC and NiO-CeO_2), the majority OR1 has the highest overlapping volume, and the minority OR2 is the second highest peak. For other materials (NiO-YSZ and CoO-YSZ), it is the minority OR2 which has the greatest overlapping volume, while the majority OR1 is the second highest peak. Finally, for the remaining materials (CoO-GDC and CoO-CeO_2), both ORs yield similar overlapping volumes. This is reasonable, since actual orientation relationships are not only influenced by the geometric coherence of the phases but also by the formation of low-energy interfaces which, in the case of DSECs, are thought to derive from a good balance of ionic charge density at the interface.[44] This was the case of OR1, considering that it shows little misfit between the ionic charge density of each phase.[41] Yet, as explained before, interface planes for the cube-on-cube orientation relationship remain unclear and are not expected to be as energetically favorable as those in the majority orientation relationship. Therefore, a very high geometrical coherence between the phases, such as the one implied by the CRLP results, may be accountable for the actual appearance of the cube-on-cube orientation relationship.

As for the impact of the chosen k parameter on the method output, it is observed that the higher the k parameter, the fewer spurious maxima are found. The choice of the optimum k parameter is not, however, straightforward. Table 3 shows the relative peak height for the three most favorable OR's determined by CRLP with k=1, 2 and 3 ($R^*=20\cdot a^*$, $r^*=0.25\cdot a^*$) for all the DSEC's studied. Despite the results being more or less consistent for all $k\neq0$ values, only the k=1 model predicts that the majority OR1 is more favorable than the minority OR2 one for all the materials (OR2<OR1). k=2 model yields this result for 4 out of the 6 materials and k=3, for 2 out of 6. On the other hand, the model with k=1 for NiO-CeO_2 and NiO-GDC found an absolute maximum which has never been observed experimentally (OR3), while k values higher than 1 resulted in the experimental ORs being the most favourable ones, as explained previously. All things considered, 2 might be the optimum k value for these particular materials, although further testing of other materials would be advisable to confirm this choice.

5. Conclusions

The CRLP method has been applied to a family of directionally solidified eutectic ceramic formed by a transition metal oxide (NiO and CoO) and an ionic conductor (YSZ, CeO_2 and GDC) that can be used as precursors for solid oxide fuel cell anodes. The only differences between the materials studied, in terms of the model, are the slightly different lattice parameters. In every case, the model has been able to predict that the two experimentally found orientation relationships are the most favourable ones according to geometric criteria.

The influence of the CRLP model parameters has been studied and a modification has been tested in order to reduce the model dependence on the reciprocal space considered, R*, to enhance the method consistency and to explore coincidences between a higher

number of families of planes in the direct lattice. The modification is based on a weighing system that attaches a higher weight and, as a consequence, more importance, to the coincidences between planes with higher interplanar spacing, i.e. those with higher atomic density.

Both the conventional and modified CRLP calculations have proved a useful tool to predict the most favourable orientation relationships in these highly textured materials. In our opinion, the ability of the CRLP model to predict, from only geometrical considerations, the actual OR's in the studied family of directionally solidified eutectic ceramics is noteworthy. According to the CRLP model, the most favourable orientation relationships between phases are those for which there are most direct-lattice planes with similar interspacing parallel to each other, i.e., those that give rise to a high intersection volume between the spheres located at the reciprocal lattices of both phases. Therefore, the favourable orientation relationship determined by the CRLP model corresponds to good 3D geometrical coherence between the adjacent crystals. From our calculations and previous experimental results, the directionally solidified eutectic ceramic systems studied are arranged in this state during the cooperative eutectic growth.

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Tables

Ceramic phase	Crystal structure	Lattice parameter [Å]
NiO	rock-salt	4.179
CoO	rock-salt	4.252
YSZ	Fluorite	5.127
GDC	Fluorite	5.418
CeO_2	Fluorite	5.411

Table 1. Crystal structure and lattice parameters of the component phases for the DSEC's studied. All phases are in the F$m\bar{3}m$ space group (reflections with mixed odd and even Miller indices are absent).

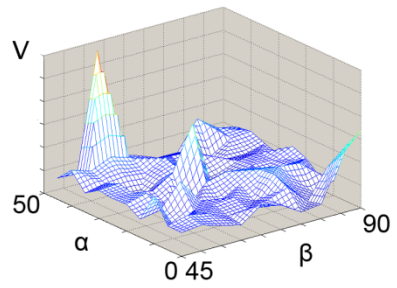
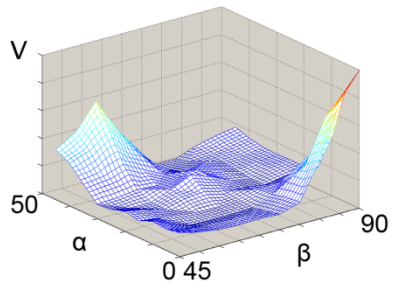
<table>
<tr><th colspan="2">Conventional vs. modified CRLP model</th></tr>
<tr><td colspan="2">$$V_{overlapping} = \sum_i V_{intersection}\left[Sphere_{hkl}^{Phase\ A}, Sphere_{h'k'l'}^{Phase\ B}\right] \cdot w_i$$</td></tr>
<tr><td colspan="2">$$w_i = \left(d_{hkl} \cdot d_{h'k'l'}\right)^k$$</td></tr>
<tr><td>Conventional
k = 0
Equal importance to all parallelisms</td><td>Modified
k ≥ 1
Enhances low-Miller-index planes parallelism</td></tr>
<tr><td> 
</td><td> 
</td></tr>
</table>

Table 2. Schematic of the differences between the conventional and the modified CRLP model. By way of comparison, the figures represent the overlapping volume calculated in a section of the Euler space with the conventional and modified methods for CoO-YSZ (R*=10·a*, r*=0.25·a*, k=0 (left) and k=2 (right)). Less spurious maxima are observed in the modified CRLP model. In addition, the results using the modified method are less dependent on the size of the reciprocal space considered (R* parameter). Thus, it is possible to explore coincidences between a higher number of direct lattice plane families.

	k = 1		k = 2		k = 3	
	OR3 / OR1	OR2 / OR1	OR3 / OR1	OR2 / OR1	OR3 / OR2	OR2 / OR1
NiO-YSZ	0.64	0.60	0.35	1.31	0.23	1.72
CoO-YSZ	0.66	0.72	0.35	1.31	0.19	1.99
NiO-CeO_2	1.10	0.85	0.96	0.68	0.55	0.82
NiO-GDC	1.11	0.85	0.98	0.67	0.56	0.80
CoO-CeO_2	0.95	0.87	0.69	0.89	0.40	1.11
CoO-GDC	0.97	0.87	0.71	0.87	0.41	1.08

Table 3. Relative peak heights of the two experimentally found orientation relationships, OR1 and OR2, and of the most favourable orientation relationship among all the others (OR3, non-experimentally-found) for k=1, 2 and 3. $R=20\cdot a^*$ and $r^*=0.25\cdot a^*$.

Figures

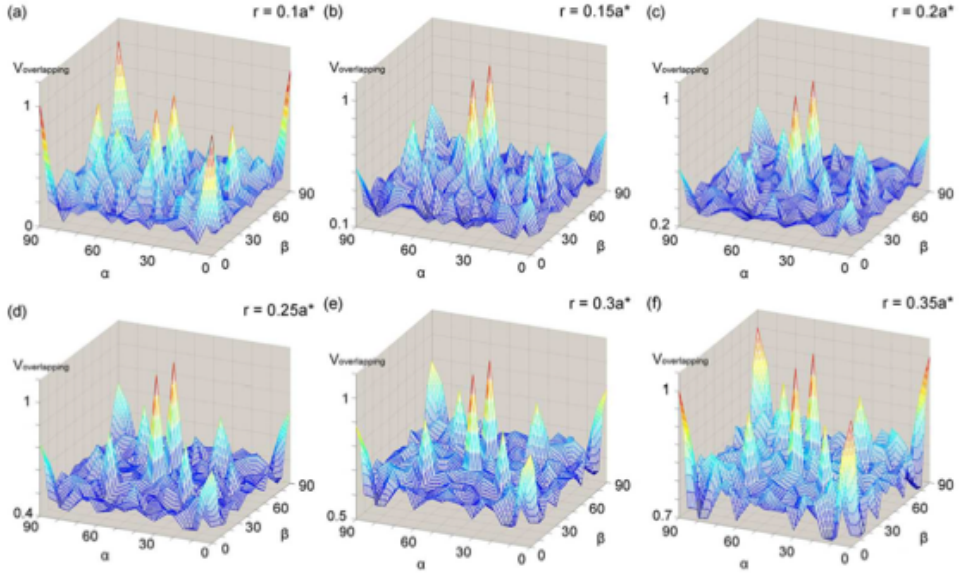


Fig. 1. Overlapping volume for CoO-YSZ as a function of α, β for r*=0.1·a*, 0.15·a*, 0.2·a*, 0.25·a*, 0.3·a*, 0.35·a*. In all cases, γ = 0º and R*=10·a*, a* being the inverse of the CoO lattice parameter.

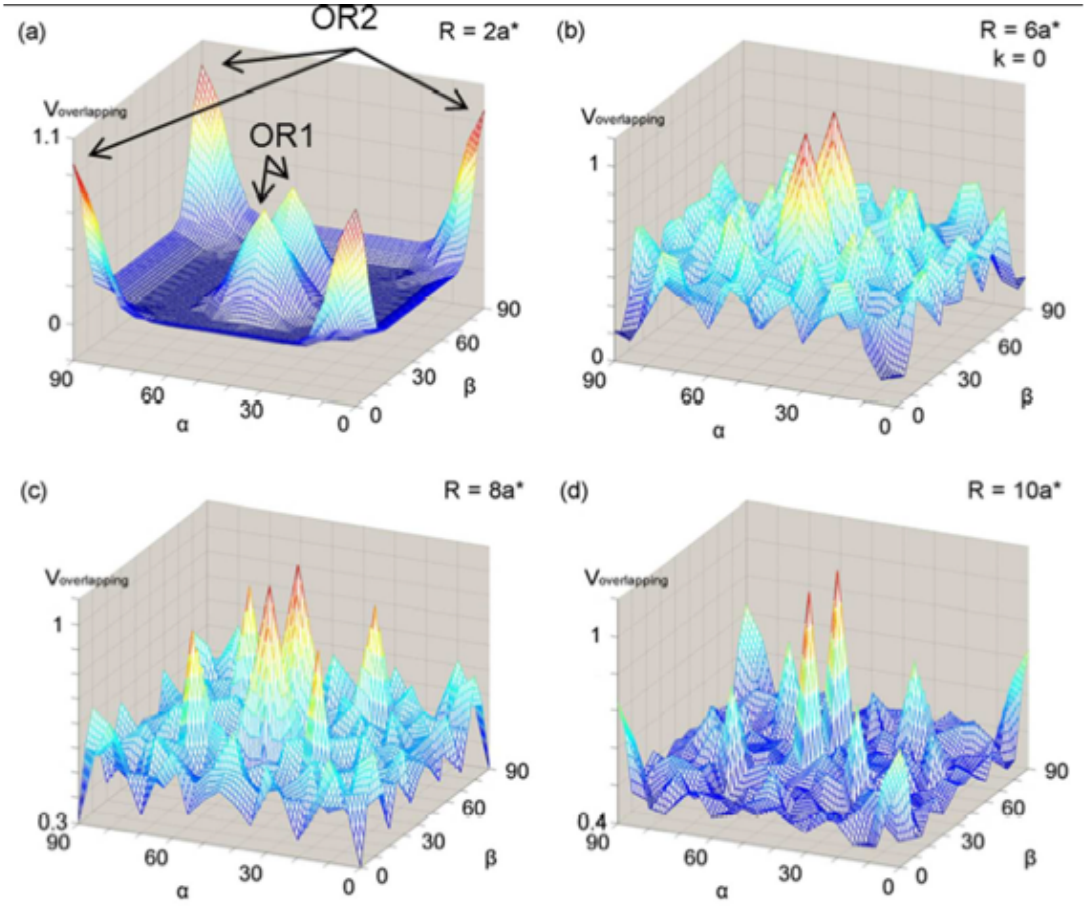


Fig. 2. Overlapping volume calculated with the conventional CRLP method (k=0) for CoO-YSZ as a function of α, β for R*=2·a*, 6·a*, 8·a* and 10·a*. In all cases, γ = 0º and r*=0.25·a*.

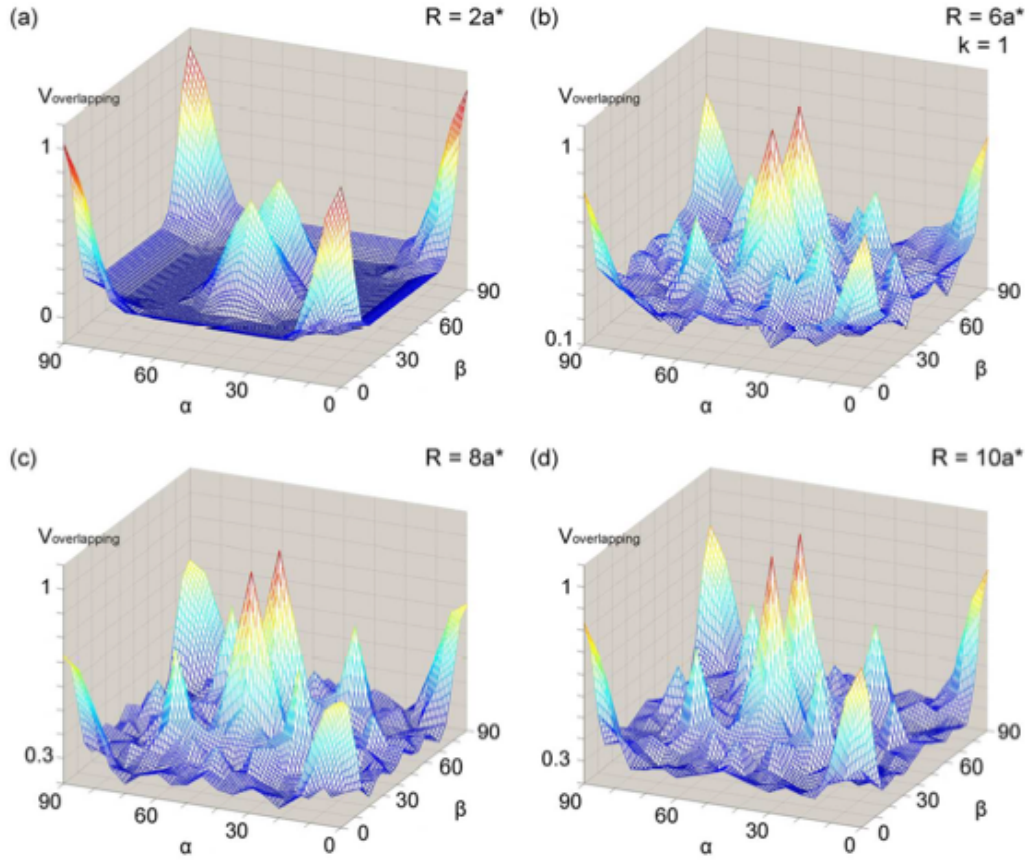


Fig. 3. Overlapping volume calculated with the modified CRLP method for CoO-YSZ as a function of α, β for R*=2·a*, 6·a*, 8·a* and 10·a*. In all cases, r*=0.25·a*, γ = 0º and k=1.

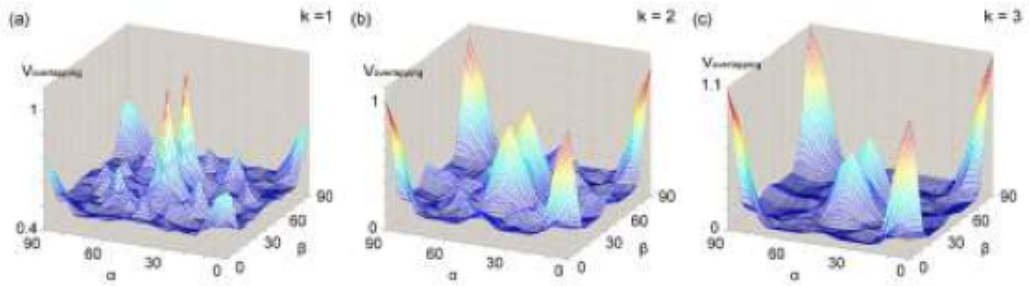


Fig. 4. Overlapping volume calculated with the modified CRLP method for CoO-YSZ as a function of α, β for k=1, 2 and 3. In all cases, γ = 0º, r*=0.25·a* and R*=20·a*.

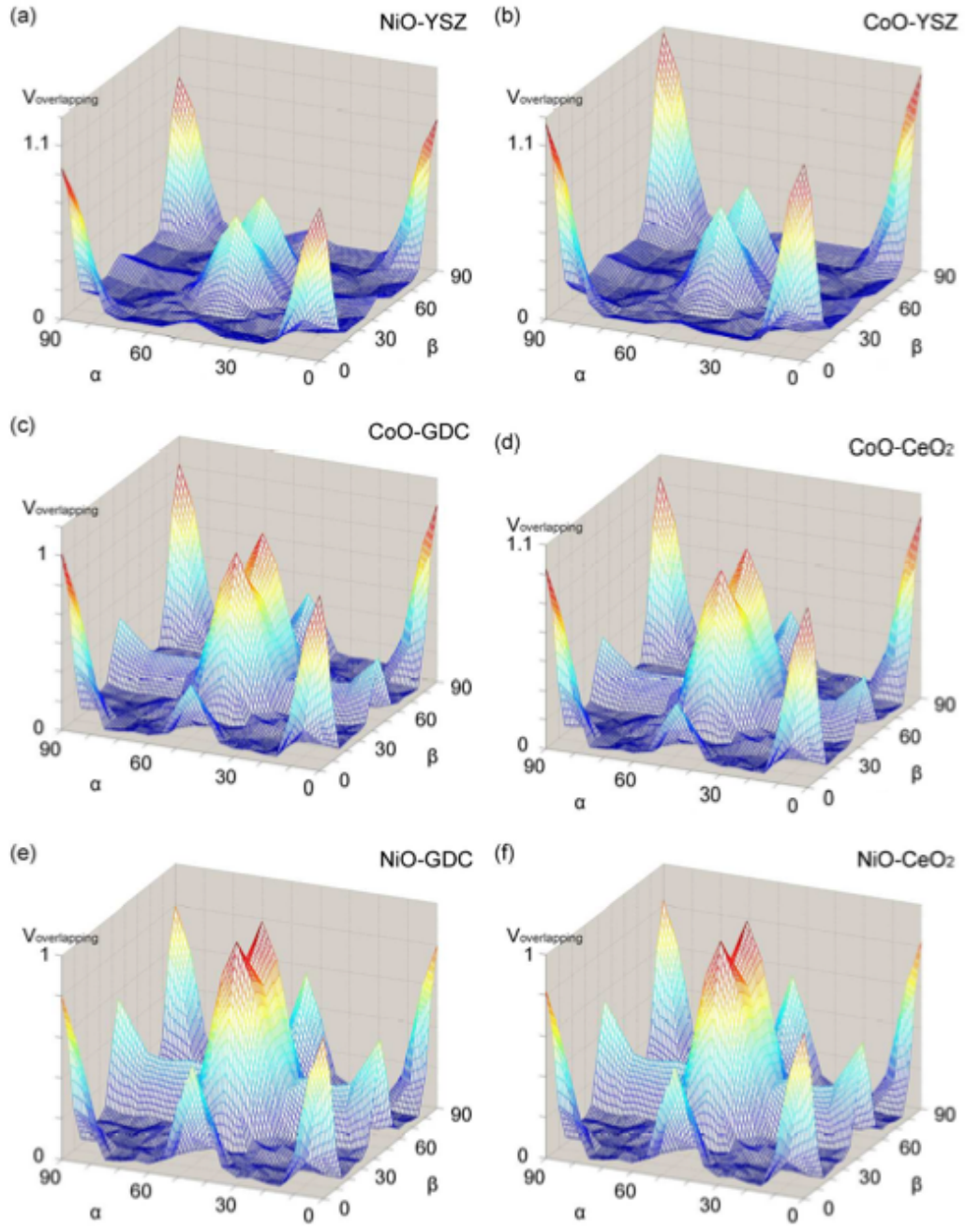


Fig. 5. Overlapping volume calculated with the modified CRLP method (k=3, R*=20·a*, r*=0.25·a* and γ = 0º) for CoO-YSZ, NiO-YSZ, NiO-CeO_2, NiO-GDC, CoO-CeO_2 and CoO-GDC, as a function of α and β.

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