

APPLICATION OF TESSELLATION TECHNIQUES IN THE STRUCTURAL ANALYSIS OF MD-SIMULATED MATERIALS

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Abstract: A new tessellation technique is applied to the structural analysis of the MD-simulated materials. The main point of the method is a suitable manipulation of the contracted simplices network. Algorithms for contraction of simplices network, as well as for radical tessellation, are presented. The contraction algorithms utilise the local atom arrangement recognition technique. An exemplary application of the new technique to MD-simulated nickel samples is described.

1. Introduction

Stochastic geometry methods are a useful technique for the study of geometrical properties of molecular dynamics (MD) simulated systems. They provide much more information about the sample structure than the analysis based only on the straightforward interpretation of angular distribution functions (ADF) and radial distribution functions (RDF), especially in the case of amorphous or multiphase materials. The most widely used techniques of the structural analysis are: common neighbours analysis (CNA) [1,2], SO₃ invariants analysis [3-7] and various tessellation techniques [8-13].

The CNA method can be used to determine the abundance of inherent structures in a sample. Each pair of atoms from the first or the second RDF peak is described by a sets of indexes. The indexes determine to which peak a given pair does belong, the number of common nearest-neighbours, the number of pairs of the first neighbours in the set of common neighbours, *etc.* The CNA is very efficient in the study of the nearest order in computer simulated amorphous materials. However, the method can give merely partial and ambiguous information about the types of crystalline phases in the analysed samples. In particular, the pairs with the same sets of indexes can belong to different phases (*e.g.* both the face-centred (fcc), and the hexagonal-close-packed (hcp) structures contain identically indexed pairs).

The SO(3) invariants analysis is based on the construction of certain sets of the SO(3) invariants for local structure (local order or bond order parameters). The comparison between such sets allows us to distinguish between some predefined reference patterns. The sets of invariants suitable for the identification of the fcc, hcp and icosahedral structures are well known [5], but the extension of the method to other geometries is rather difficult: due to a limited number of invariants the conclusions about the local structure are not necessarily unambiguous. A similar method was used for the investigation of a non-local order [3]: the bond order parameters were constructed for all the atoms in the sample, making it possible to draw out some conclusions about the orientational order.

The tessellation method consists in the division of the total volume of the simulation box into an array of subvolumes belonging to the atoms. A number of various constructions is known. The most difused one is the partition into the Voronoi polyhedra (VP). VP is defined as the minimal polyhedron whose planar faces bisect at right angles the lines joining an atom to its neighbours; a pedantic definition is given by Brostow and co-workers [11]. However, one can use more general tessellation, based on radical planes [9]. In this case each atom from the array is characterised by an additional parameter, which can be interpreted as the atom radius. Vertex of a radical polyhedron (RP) is such a point in the space, that the vertex-tangent distances to four neighbouring atomic spheres are equal. It is obvious that when all atomic radii are equal, the radical-plane tessellation is identical to the Voronoi partition. Thus, all the properties of RPs are valid for the VPs. The RP diagram (alternatively RP network or RP graph), *i.e.* the set of RPs constructed for all the atoms in the sample, is rigorous; it splits in a unique manner the total sample volume into the zones owned by each atom. Radical simplices (RSs) are geometrically dual polyhedra to RPs; that is, a vertex of an RP is the central site of the corresponding RS, and each atom (centre of a RP) is a vertex of the corresponding RS. One can assign each vertex of the former lattice to the elementary units of the latter one. The faces of RSs intersect the edges of RPs, and the faces of RPs intersect the edges of RSs. In the case of the Voronoi partition the simplices are called Delunay simplices (DS). RP and RS (VP, and DS, respectively) networks contain a full information about the structure of the analysed sample.

The purpose of this paper is to present some applications of the tessellation methods for the analysis of geometrical properties of MD-simulated materials. Since contemporary computers allow to carry out the MD simulations for systems composed of hundreds thousands of particles, one needs an efficient tessellation algorithm (Section 2). The methodology of the networks treatment, developed here, differs from the ones presented earlier by other authors, where each RP in the sample was constructed and analysed independently from other RPs. Thus, each vertex of the RP network was constructed four times (in the non-degenerated case). Our new algorithm determines each vertex only once. In Section 3 we describe the algorithm of the RP network contraction. The contraction eliminates an influence of atomic position perturbation of the network. These perturbations originate from thermal motions or

can be totally incidental (finite precision of the coordinates storage). Section 4 contains some examples of application of the technique, and Section 5 concluding remarks.

2. Tessellation algorithm

As it was mentioned in the introduction, the RP network is constructed in such a way that each vertex is determined only once. To make our algorithm more general, the array of atoms is decomposed into two arrays: the *centres array*, and the *neighbours array*. The centres array contains the atoms, for which RPs are to be determined. The neighbours array contains all the neighbours of the atoms of the centres array. The neighbours array can include atoms of the centres array. For instance, in SiO₂ sample we can chose Si atoms as centres and all atoms as neighbours. Thus, we determine the RP for Si atoms only. The geometrical neighbours of Si atom could be either other silicon atoms, or oxygen atoms. If the two arrays are equal, we deal with the classical partition.

Let us introduce some auxiliary definitions, usefull in our description of the construction algorithms of the networks. An *open centre* is a centre that is actually not contained in any of just constructed simplices. Two simplices are *contiguous* if they share a common face. Let *open simplex* be a simplex that has no contiguous simplices in the set of already constructed simplices. An *open face* is a face of a simplex, that contains some centres and the face is not shared with any simplices already constructed (all the faces that do not contain any centre are never open). The algorithm can be summarised as follows:

1. Create list of *open centres*.
2. Choose an *open centre* C form the *open centres* list. If the *open centres* list is empty, go to step 7, else go to the next step.
3. Construct the *first simplex* that contains C and three atoms from the *neighbours array* (an exact description of the construction is given below). Add this simplex to the simplices list. Remove C from the *open centres* list.
4. Choose simplex S which has an *open face*. If such a simplex does not exist, go to step 6, else go to the next step.
5. Construct a new simplex *contiguous* to S (for an exact description see below). The simplices must share the *open face* considered in step 4. A new fourth atom of S must be taken from the *neighbours array*. If it belongs simultaneously to the *centre array*, remove it from the *open centres* list. Add S to the simplices list, and go to step 4.
6. Go to step 2.
7. The end of the RS construction procedure.

Steps 2 and 4 do not require any comments. The construction of the *first simplex* and the next simplices in steps 3 and 5 is more complicated. In our realisation we adopted the routine proposed by Tanamura and co-workers [10]. Since their algorithm was written only for the Voronoi tessellation, we have generalised it on the case of

the radical tessellation. The changes have been made in all the steps, were the distances between the atoms, and the vertices or the positions of faces, are calculated. The suitable modifications base on the equation for the RP network vertices given by Gellatly and Finney [9]. At this stage it is convenient to introduce a routine of the degeneration detection, making the algorithm more stable. Degeneration occurs when one can construct more than one simplex (by adding an atom to three atoms defining simplex face) that have the same RP vertex coordinations. It is possible to construct an algorithm that can work with degenerated systems, but because the degeneration does not occur in practice, it would be computationally inefficient. In the case of degeneration we simply disturb the positions of atoms by a small fraction of the nearest-neighbour distance (of order 10^{-5}), and then we start the procedure from the beginning. The positions of atoms are restored after the procedure ends. In view of the considerations of Section 3, the perturbation of atomic positions does not have any influence on the final conclusions about the structure of the analysed sample. Realisation of step 5 is similar to the last step of the construction of the *first simplexes* (cf. [10]). We know the face (in this case it is an *open face*) and we have to build a new simplex by adding an atom to this face. Care must be taken at selection of candidates for this fourth atom. We have to choose an atom which is placed on the opposite side of the face (in respect to the fourth atom of the simplex from step 4). After executing steps 1-7 described above we have the complete list of simplices of the analysed sample. If *centres array* and *neighbours array* are identical, we have a full RS graph, else we have only its subgraph. In both cases, we can determine the set of radical polyhedra for all centres, utilising only information contained in the simplices list.

The determination of the RS graph is only a preliminary step of our tessellation method. At this stage the RPs are rather complex polyhedra, containing many faces and vertices. Their topology does not allow to draw out any unambiguous conclusion on the structure. It is clear that the basic information about the local structure at any atom is carried mainly by big faces and long edges. The small faces and short edges have only inconspicuous meaning. To utilise the RPs efficiently we contract short edges and small faces of the RP graph.

To motivate the need of the contraction, let us consider the influence of small perturbations of atomic positions in an arbitrary crystalline lattice on their RP networks. A characteristic feature of certain crystalline networks (e.g. fcc or hcp) is the existence of degenerate vertices and edges in their RP graphs. Degenerate neighbours corresponding to such vertices or edges have been defined by Brostow and co-workers [11]. A degenerate vertex is common to more than four edges, while a degenerate edge is common to more than three faces of the RP graph. As discussed previously [11], we also have so called indirect neighbours: there is a common face, but the midpoint of the line connecting the atoms does not belong to that face. If the midpoint belongs to the common face, we have the simplest case of direct neighbours. Here, direct and indirect neighbours will not be distinguished, and they will be called geometric neighbours. It is obvious that an arbitrary small displacement of atoms in

the crystalline structure removes the degeneracy. In the place of a degenerate vertex, a small face or a short edge will appear, and degenerate edges will become small elongated faces. The local arrangement is practically the same in perturbed and in unperturbed structures, but the topology of radical polyhedra is much more complicated in the former case. Thus, eliminating short edges and small faces from the VP network (by contracting them to vertices or edges), we remove the effects due to the perturbations and simplify the polyhedra topology. The same result could be achieved by suitable displacements of the atoms. However, since we have no information about the individual fluctuations of the atoms in the sample, such a procedure cannot be realised in practice. In a structure in which the degenerate vertices are absent (*e.g.* bcc lattice), a small perturbation of the position does not change the topology of the network. Thus, the analysis of such structures can be performed going directly to the next stage of the procedure, described in Section 4.

3. Graph contraction algorithm

To proceed with the graph contraction one needs to find the set of edges that are to be contracted. This can be done directly by computing all edges lengths and selecting all the edges shorter than a certain predefined threshold value. The threshold can be determined on the basis of the edge length distribution. The typical edge length distribution for the face centred structure is presented in Figure 1. As it is seen, it is difficult to establish the threshold value, because the large peak of longer edges can not be separated from the peak of shorter edges. To avoid the difficulty we propose an algorithm which is able to analyse sub graphs of the RS network. Each of simplices belonging to any of these subgraphs should contain a specified central atom. Contraction of the whole RS network is performed subsequently in each subgraph. After

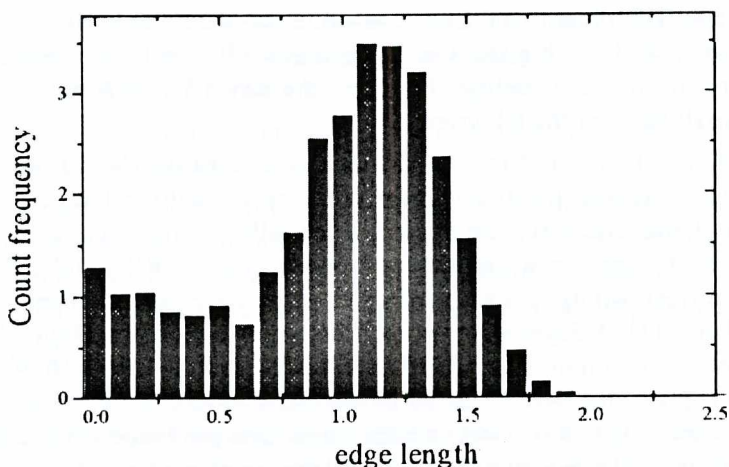


Figure 1. Edge lengths distribution for the Voronoi graph of a perturbed fcc structure. The ideal nodal positions are shifted by 0.15 of the nearest neighbour distance in random direction. Edge lengths are measured in the unit of the average edge length.

each step the RS network is updated, and the subgraphs are recalculated. The algorithm can be summarised as follows:

1. Take centre C from the *centres list*, C not considered previously. If it is not possible go to step 7.
2. Select subgraph S of the RS network. All simplices from the subgraph must contain C . With no degeneration the set of atoms constituting S is an atomic polyhedron related to C . Else, it contains in addition all degenerated neighbours.
3. Determine radical polyhedron P of centre C , using the subgraph S (in this step care must be taken in the selection of vertices, because not all vertices associated with simplices from S are included in P ; see the text below). Evaluate edge lengths of this polyhedron.
4. Select set E of the edges of P , that are not contained in the previously contracted polyhedra. It can be done simply by checking a composition of the simplices which define the vertex of the edges. If the simplices associated to both vertices of the edge contain the centre considered previously, the edge is not being selected.
5. Contract subgraph S , using list E (for exact description see below), and update the RS network.
6. Go to step 2.
7. The network is contracted. End of the procedure.

Some of the above steps should be elucidated. In step 3 we determine a polyhedron using a subgraph of the RS network. This can be done by examining the geometrical relations between the simplices from S . The vertices associated to these simplices constitute the polyhedron. Two mutually *contiguous* simplices in S define the edges of a polyhedron. The faces are defined by a set of simplices that contain the central atom, and its geometrical neighbours. Because the RS network is now after several contractions of the subgraphs determined for other centres, it is possible that some vertices on the polyhedron constructed in such a way are topologically incorrect. There may occur vertices in which only two edges meet. Topologically correct situation is when three and more edges of a polyhedron meet in a vertex. Taking it into account one must reconstruct the polyhedron removing all the incorrect vertices and change the geometrical relation between the simplices. The simplices that are contiguous to incorrect vertex became contiguous to each other, but only for the moment in which the polyhedron is being constructed. In step 5 we utilise our previous technique of Voronoi polyhedra contraction [14]. The algorithm applied here allows for a very careful contraction of subgraph S , by an appropriate selection of contracted edges and permanent control of polyhedron shape during the process. Let us itemise the main points of this technique:

1. For the RP under consideration contract all the edges shorter than a certain fraction x of the average edge length.
2. Find the shortest edge.

3. If the edge is shorter than a fraction y ($y > x$) of the average edge, check the shape of the polyhedron (for description see below). If the shape belongs to the set of the predefined patterns, take the next polyhedron, and go to step 1; otherwise contract the edge under consideration, and go to step 2.

4. If the edge is longer than a fraction y of the average edge end the procedure.

Detailed tests of the algorithm efficiency accomplished for monatomic metallic MD-simulated samples allowed to establish the optimal values of the parameters x and y to be 0.3, and 0.5, respectively. $x < 0.3$ results in switching on the shape recognition procedure sooner; if the shape of a given polyhedron is undefined, there are no consequences except the slowing down of computations. On the other hand, $y > 0.5$ for an undefined shape leads to too many contractions, leaving eventually a polyhedron with only few edges and faces.

Let us turn to the problem of the polyhedron shape recognition. The shape of an arbitrary polyhedron can be described by certain sets of integers, as $F = (f_3, f_4, f_5, \dots)$, $V = (v_3, v_4, v_5 \dots)$, $E = (e_4, e_5, e_6, \dots)$. Here f_i is the number of the i -edged faces in the polyhedron; v_i is the number of the vertices, from which exactly i edges originate. In the case of a non-degenerated VP, only v_3 do not vanish, hence $i - 3$ equals to the degeneration degree. Finally, e_i is the number of edges for which i equals to $j + k + 4$, where j and k are the degrees of degeneration of both vertices associated to the edge. It is not complicated to introduce more sets like these (e.g. in the form of two or three dimensional array), but our experience shows, that the sets F , V , and E , defined above suffice to perform an efficient structural analysis. Two polyhedrons are said to have the same topological structure if they have the same F , V , and E sets. Thus, we can compare all the constructed polyhedra with an arbitrary set of reference polyhedra. For instance, the fcc polyhedron has $F = (0, 12)$, $V = (8, 6)$, $E = (0, 24)$, and the hcp polyhedron has $F = (0, 12)$, $V = (8, 6)$, $E = (3, 18, 3)$.

At this stage one can use the contracted network to establish geometrical properties of the analysed structure in many various ways. A particular realisation depends on the questions one asks. In the subsequent section we show some applications of our method.

4. Exemplary application of the technique

Let us concentrate on the geometrical properties of individual elementary units of the RS, and RP networks, as well as on spatial correlations between the units. Elementary units of the RS network are radical simplices. In general, they are rather simple polyhedra, and in order to recognise their shape it is enough to count the polyhedra-forming atoms. In the case of four-atom simplices we can measure their shapes using the Medvedev and Naberukhin [15] tetrahedrality parameter T ,

$$T = \frac{\sum (l_i - l_j)^2}{7^2}$$

where l_i is the length of the i -th simplex edge, and is the average edge length of the simplex. Spatial correlations can be investigated easily by analysing the properties of the cluster of simplices connected by a face, a vertex, or an edge (by a cluster we mean the connected subgraph of the graph). Elementary units of RP networks are radical polyhedra. Their shapes are much more complicated than of the RS's, but we can still utilise the shape recognition technique of the previous Section, defining a sufficiently large set of the predefined patterns. The cluster structure provides an immediate evidence of the existence of distinct crystalline phases in an MD-simulated material.

We will apply the described techniques to the study of two MD-simulated nickel (Ni) samples. In both cases simulation box contains 500 atoms interacting by a model potential based on tight-binding calculations; the parametrisation has been performed using experimental cohesive energy, bulk modulus and elastic constants as constraints [16]. The simulations were performed in the microcanonical ensemble. The samples were prepared initially as well equilibrated melts at 5000 K and density 8500 kg/m^3 (the samples are slightly rarefied, density of nickel at normal conditions is about 8908 kg/m^3). The liquids were equilibrated for 10^5 time steps ($\Delta t = 10^{-15} \text{ s}$). The first sample has been quenched directly to 1 K (fast cooling). The second one has been cooled down to 1 K passing the equilibrium states at 2500, 1250, 600, 300, and 150 K (slow cooling).

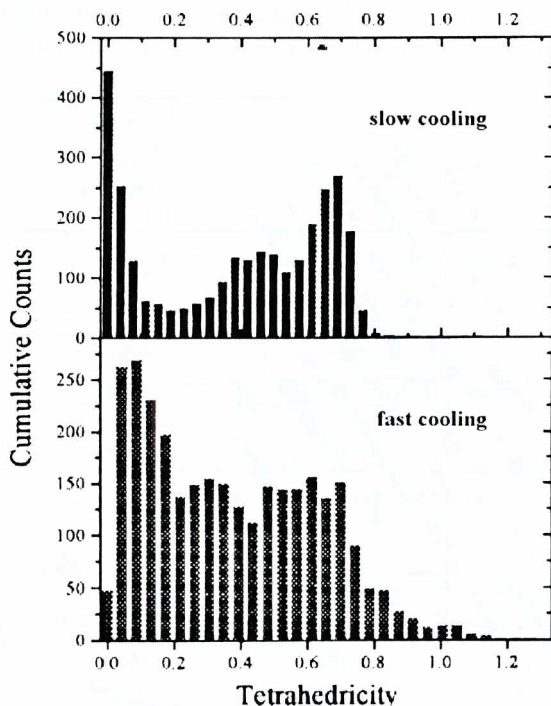


Figure 2. Distribution of tetrahedrality parameter.

We deal with a monatomic system (all atomic radii are equal), thus the radical tessellation is equivalent to the Voronoi partition. The simplices network has been contracted with the parameters $x = 0.1$ and $y = 0.4$ (see Section 3). The distribution of the tetrahedrlicity parameter in both samples is shown in Figure 2. More detailed analysis reveals that only the simplices with a large value of the tetrahedrlicity parameter are converted into degenerated units during the contraction of the RS network (formally, a simplex is a polyhedron composed on $D+1$ vertices, where D is the dimension of the space; in three dimensional space only disordered tetrahedra can be called simplices; but here we will use notion simplices to describe all elementary units of RS network underlining their origin). Statistics of the simplices size for both samples is presented in Table 1. It is seen that the number of simplices is about 10% higher in the quickly cooled sample than in the slowly cooled one. In both cases the RS networks are composed mainly of four-atom simplices, but also a significant amount of the octahedral RS network units is detected. This can mean that the sample contains certain zones of fcc or hcp ordering. Because none of monatomic ideal crystalline networks contains 5-atom simplices, their appearance in the samples results from a local crystallisation. Tables 2 and 3 contain the cluster properties of 4-, and 6- atom simplices, respectively. In the fast cooling case almost all 4-atom simplices form a single cluster, exactly as in the rcp or bcc structures. Figures 3 and 4 present spatial distributions of 4-, and 6-atom simplices in slowly cooled sample. Points are placed in the centres of the simplices, and sticks between them indicate that they share a face. As it is seen, the clusters have an arrangement that does not appear in the quickly cooled sample, where the whole simulation box is filled by a

	simplex sizes		
	4	5	6
slow cooling	79.4%	4.9%	15.6%
fast cooling	77.7%	14.8%	7.3%

Table 1. Simplex size statistics in slowly and quickly cooled samples, containing 1973, and 2114 simplices, respectively.

	number of clusters	number of isolated simplices	number of simplices in couples	size of the biggest cluster
slow cooling	24	6.1%	19.1%	74.3%
fast cooling	32	1.4%	0.6%	97.6%

Table 2. Properties of clusters composed of 4-atom simplices.

	number of clusters	number of isolated simplices	number of simplices in couples	size of the biggest cluster
slow cooling	110	9.7%	15.6%	2.7%
fast cooling	82	30.6%	24.2%	6.4%

Table 3. Properties of clusters composed of 6-atom simplices.

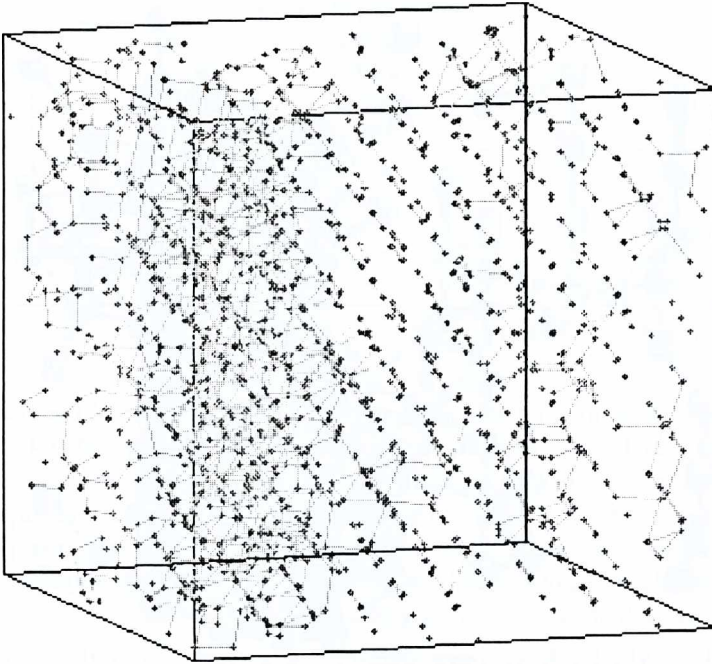


Figure 3. Spatial distribution of 4-atom simplices in the slowly cooled Ni sample. Points indicate the centres of the simplices, and sticks join the face-sharing simplices.

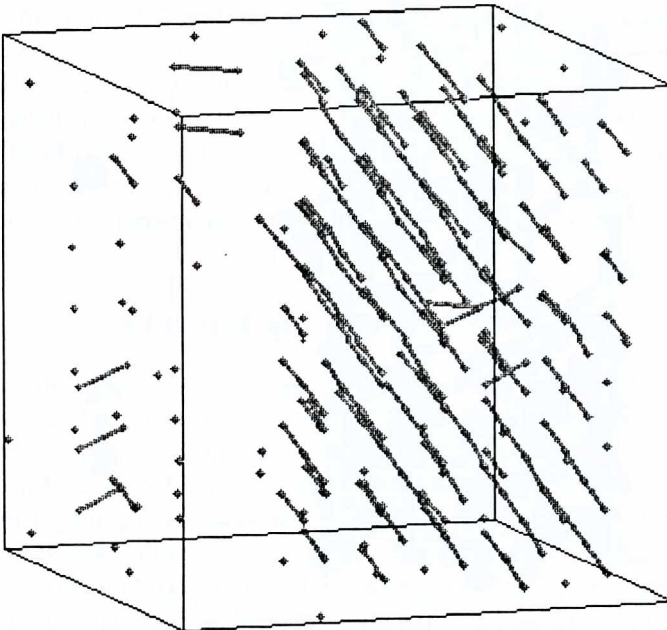


Figure 4. Spatial distribution of 6-atom simplices in the slowly cooled Ni sample.

	number of polyhedra	number of faces
slow cooling	2.8%	11
	45.4%	12
	45%	13
	6.8%	14
fast cooling	6.6%	11
	31.2%	12
	36.2%	13
	20%	14
	5.2%	15

Table 4. Numbers of faces in the polyhedra.

single cluster of 4-atom simplices. It is evident that the volume of the slowly cooled sample is occupied by two distinct phases. One of them is composed of 4-, and 6-atom simplices. 4-atom simplices appear mainly in pairs, whereas the 6-atom simplices form much longer linear clusters. The pairs and the linear clusters have the same orientation. Exactly the same properties characterise the hcp structure. The second of the detected phases contains only 4-atom simplices, and the following analysis of the RP network will show that this is a bcc phase.

Radical polyhedra have been determined directly from the contracted RS network. Statistics of numbers of faces in the polyhedra is collected in Table 4. In the slowly cooled sample more than 90% of atoms have 12- or 14-faced polyhedra, whereas in the quickly cooled sample, polyhedra with 13 faces are most frequent. Using the shape recognition technique we can say that 31% of atoms in the slowly cooled sample have the first co-ordination shells as in a hcp crystal, 42% as in a bcc crystal, and only 6.2% as in a fcc crystal. The applied technique does not give so good results in the quickly cooled sample. Almost 95% of atoms have unrecognised neighbourhood structure, and 3% of them have an icosahedral local arrangement. Figures 5 and 6 present spatial arrangements of bcc-, and hcp-surrounded atoms in the slowly cooled sample. The Figures confirm our former conclusions about the existence of the hcp and bcc phases in the sample.

5. Concluding remarks

In this contribution we have described an effective way of utilising the tessellation method in structural analysis of MD simulated materials. Each radical diagram constructed for any sample contains some excess information concerning meaningless fluctuation of atomic position. In order to extract only useful data, we have applied a suitable graph contraction technique. The main point of the contraction algorithm is a careful removing of the short edges from the radical graph. We have achieved this by subsequent contractions of the subgraph related to each central atom. Proceeding in this way we have had under control an essential criterion of the short edges selection. It depends on the local subgraph structure, whether an edge is qualified as short or not. We have applied our technique in the analysis of structure

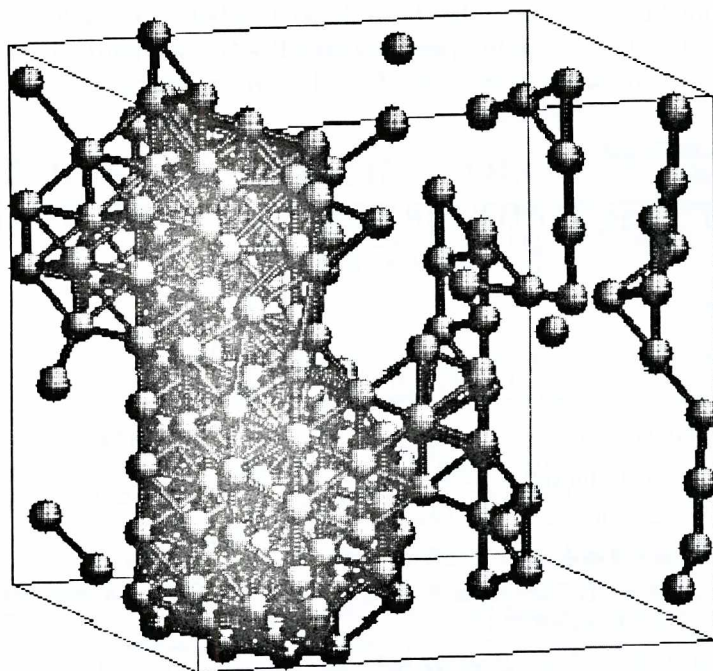


Figure 5. Spatial arrangement of bcc-surrounded atoms in the slowly cooled Ni sample.

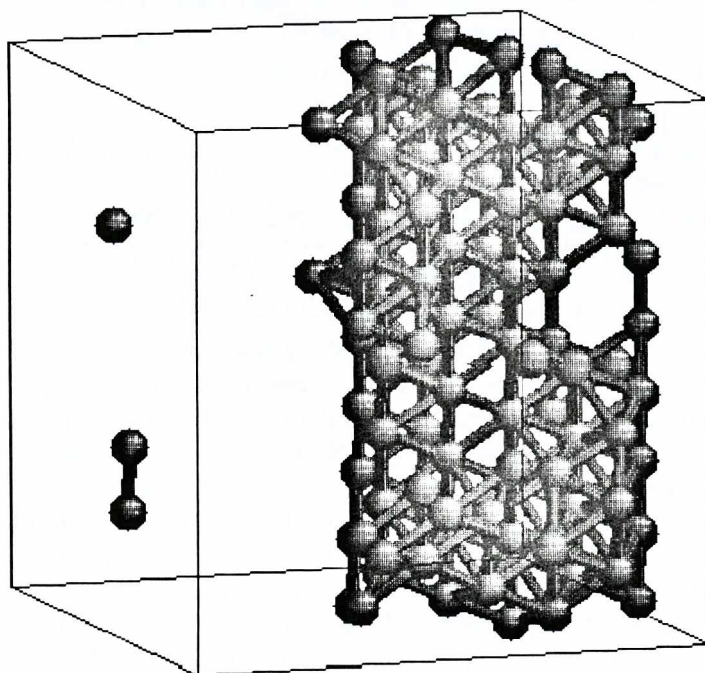


Figure 6. Spatial arrangement of hcp-surrounded atoms in the slowly cooled Ni sample.

slowly and quickly cooled Ni. It has been shown that both RS and dual RP networks contain a lot of useful information, and that the RP network is particularly convenient to perform the recognition of the type of local atom arrangement.

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