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Parallel algorithm of solidification process simulation for large-sized system of liquid metal atoms①

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Abstract: A parallel arithmetic program for the molecular dynamics (MD) simulation study of a large-sized system consisting of 50 000－100 000 atoms of liquid metals is reformed, based on the cascade arithmetic program used for the molecular dynamics simulation study of a small-sized system consisting of 500－1 000 atoms. The program is used to simulate the rapid solidification processes of liquid metal Al system. Some new results, such as larger clusters composed of more than 3－6 smaller clusters (icosahedra or defect icosahedra) obtained in the system of 50 000 atoms, however, the larger clusters can not be seen in the small-sized system of 500－1 000 atoms. On the other hand, the results from this simulation study would be more closed to the real situation of the system under consideration because the influence of boundary conditions is decreased remarkably. It can be expected that from the parallel algorithm combined with the higher performance super-computer, the total number of atoms in simulation system can be enlarged again up to tens, even hundreds times in the near future.

Key words: parallel algorithm; liquid metals; solidification processes; large-sized system; computer simulation

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1 INTRODUCTION

It is well known that in the study of microstructures of materials, sometimes it is difficult to get the desired results because of the limit of experiment conditions. For example, the X-ray diffraction cannot be used to show the real microstructures of liquid or amorphous metals within their short-range order. During the rapid cooling processes from liquid to glass metals, it is also difficult to observe the transient change of their microstructures by instruments, and so on. With the development of computer science and technology, however, those results always can be easily acquired by computer simulation methods, so the computer simulation has been considered as an important path to study the microstructures of materials. There are many different kinds of computer simulation methods, of which the molecular dynamics (MD) simulation is rather suitable to study the dynamic solidification processes of liquid and amorphous metals. We have focused on this aspect early in 1990s, up to now the MD method is still an important method to study the microstructures of liquid and amorphous metals. However, using the MD method by middle-sized or micro-computers, owing to the limit of memory and computing velocity, we can only perform the simulation of small-sized systems consisting of 500－1 000 metal atoms. Thus the simulation results would be greatly different from the realities of the system, although they have revealed many microscopic characteristics and rules of materials. It can be predicted that as long as we can simulate a larger system on a super-computer, the results will be closer to the realities of the system and will have more important instructing meaning to the productive practice. However, a simulation calculation of a larger system by the super-computer should adopt the corresponding parallel algorithm to bring the advantages of the supercomputer into full play. Based on previous works[1－5], therefore, this paper will try to develop the initial cascade program of molecular dynamics simulation by a parallel program to enlarge the amount of the simulating system up to 50 000－100 000 atoms. Thus the goal of the simulation results being closer to the realities of system will come true.

2 ALGORITHM ANALYSES

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Molecular dynamics, as we know, is an important method to solve the classical motion equations of systems comprising many particles having interacting forces between each other. Once the initial position (coordinate r) and speed (v) of each atom in the systems are given, and the interacting potential $V(r)$ between atoms also known[6,7], the forces on each atom can be calculated, and the new position and speed will be obtained. Thus, by tracing this calculation step by step, the dynamic situations of the system can be simulated, and the instantaneous microstructures of the system can be got. From these results, we can also calculate the macroscopic thermodynamics properties of the system.

In MD simulation, the Verlet[8,9] algorithm is often used to solve the classical motion equations of particle systems by computer. This algorithm is rather mature and widely used, and we have got many simulation results of small-sized systems consisting of 500 - 1 000 atoms by it. Hence, this paper just tries to introduce the parallel algorithm into the original cascade algorithm but not develop an entirely new algorithm for parallel computing simulation. This will make it suitable to compare the simulation results from a large-sized system with those from a small-sized system. According to Verlet algorithm, once the coordinates and speed of each atom at a certain time (t) have been given, then at time ($t + \mathrm{d}t$), we will have

$$r(t+\mathrm{d}t) = 2r(t) - r(t-\mathrm{d}t) + \mathrm{d}t^2 a(t) \quad (1)$$

Where the speed is not necessary to be calculated, but if we want to estimate the kinetic energy, the speed v should be got by the following equation:

$$v(t) = [r(t+\mathrm{d}t) - r(t-\mathrm{d}t)]/2\mathrm{d}t \quad (2)$$

In Eqns. (1) and (2), the calculation of the acceleration a of each atom is equivalent to the calculation of the force f on the atom. We adopt the pair potential approximation to calculate the force f. The force on atom i can be calculated by the following equation:

$$f_i = \sum f_{ij}(t), \quad i \neq j \quad (3)$$

where f_{ij} is the acting force on atom i by atom j.

From Eqns. (1) and (2), it can be easily seen that, theoretically speaking, the time consumption for calculating the forces will be proportional to the square of the number of atoms in the simulating system. Thus the part of the time consumption for calculating forces will occupy the most proportion of the whole calculation, and the ratio will increase with the increase of the number of atoms in the system. When the number of atoms is as great as 50 000, the time consumption is unacceptable, thus the total number of atoms we can simulate has been limited.

In the cascade program used on microcomputer, we have saved the calculating time by sacrificing computers' memories. According to this method, we record the called neighbor atoms those within the distance less than RCUT (distance, at that the pair-potential is cut off) to a given atom in a neighborhood list at time t (the other atoms whose distances more than RCUT can be considered to be omitted, namely their pair-potential will close to a constant). Normally we use a large array to store all the neighbor atoms in the neighborhood lists of each atom. At the next time step ($t + \mathrm{d}t$), if the displacements of all the atoms are not over a limit value, it is considered that the neighborhood atoms around each atom would remain the same neighborhood atoms recorded at time t. In this case, the program calls subroutine F2 to calculate the forces on each atom. If the above conditions cannot be satisfied, the calculation of the forces on each atom should consider the effects of all other atoms. The program will call subroutine F1 to calculate the forces on each atom, at the same time, the neighbor atoms surrounding each atom would be changed and the neighborhood lists should be updated. Because the number of neighborhood atoms around an atom will not be changed with the increase of the total number of atoms in the system, the time-consumption of F2 will be directly proportional to the total number of atoms. The F1 is required to calculate the interacting forces between all the atoms in the system, thus its time consumption will be directly proportional to the square of the total number, as shown in Table 1.

Table 1 Time consumption of original algorithm

	Time consumption of each circle/s				
Number of atoms in system	500	1 000	2 000	5 000	5 000 (Predicting)
Subroutine F1	0.43	1.536	5.583	32.90	3 000
Subroutine F2	0.264	0.605	1.2 012	3.115	31

Using this method, some time consumption can be saved, but a great deal of memory must be occupied to store the neighborhood lists. Since the number of neighbor atoms around each atom is about 300 - 600, normally we define a very large array LIST (NS, 600) to store the lists, here NS is the total number of atoms in the simulating system. Thus when simulating large systems, for example, when NS= 50 000, the memory required for the array will be 50 000* 600* 4/1024/1024= 114.44 M bytes, adding to other memory requires, the memory of a simple computer is difficult to meet the need. Accordingly the neighborhood lists method cannot be used when NS is equal to or even larger than 50 000, which means that at this time the F2 cannot be used, all of the works for calculating forces must be done only by F1. However, the time consumption for this calculation is not acceptable.

Consequently, we must consider using super-

computers and parallel algorithm to achieve our goal of simulating the large-sized systems comprising 50 000 atoms. Based on the original cascade algorithm, we present the following parallel algorithm[10]. The parallel algorithm for calculating force is shown in Fig. 1. The master-slave type is adopted in parallel computing model. We divide all the atoms in the system into each slave machines in average, including the coordinates, neighborhood list of atoms. For example, the whole system comprises 50 000 atoms, and we prepare to divide them into 50 slave machines. So, the No 1 to No 1 000 atoms are divided into No 1 slave machine, while No 1 001 to No 2 000 atoms into No 2 slave machine, and so on. When the main program calculates the forces, the current coordinates of atoms will be broadcast to every slave machines. Then each slave machine calculates the forces on each atom being divided into this machine. As in the above example, the No 1 slave machine is responsible for calculating the forces on No 1 atom to No 1 000 atom, and the No 2 slave machine for calculating the forces on No 1 001 to No 2 000 atom, and so on. When each slave machine finished its calculating, they will send the results (the forces) back to master machine, and the next step will be continued. In this way, the calculation of forces will be parallel and the speed will be greatly accelerated. What is more, the gigantic neighborhood list array has been divided into each slave machine, thus this array will be handled by each slave but not by master. Finally, the simulation of large-sized systems will not be limited due to the memory and the computing speed of computers.

The advantages of adopting the parallel algorithm are as follows.

1) The new algorithm has not been different from old algorithm essentially, only the parallel parts being introduced into it. The simulation results obtained should be the same as original, so as to detect and compare the results from both algorithms.

2) This algorithm adopts master-slave parallel model, being clear and obvious, and easy for maintaining and adjusting the number of slave machines.

3) This algorithm has better solved the memory problem, divided the great number of memories needed for the simulation of large-sized systems into each slave machine. As long as the number of parallel machines, the memory and computing speed of computers are enough, the number of atoms in simulation system can be increased with hundreds and even thousands times.

The theoretical accelerating ratio of the above algorithm is R1* C/2, here C is the number of slave machines, R1 is a coefficient meaning the proportion of the part of calculating forces to the whole calculation. When the total number of atoms in the system is very large, the value of R1, in general, is above 90%. In addition, the reason why the above expression divided by 2 should be that the interacting forces between two atoms are relative each other. Each force is only calculated one time on single computer, however, it has been calculated two times in parallel algorithm, namely, there is one time to be repeated.

3 ALGORITHM REALIZATION

The program processed by parallel algorithm has been run steadily on the YH-3M supercomputer designed and produced by our country, adopting PVM programming with master-slave type. The master will run on the front workstation of YH-3M, the slave will be given to the processing machines of YH-3M. The two subroutines F1 and F2 in the

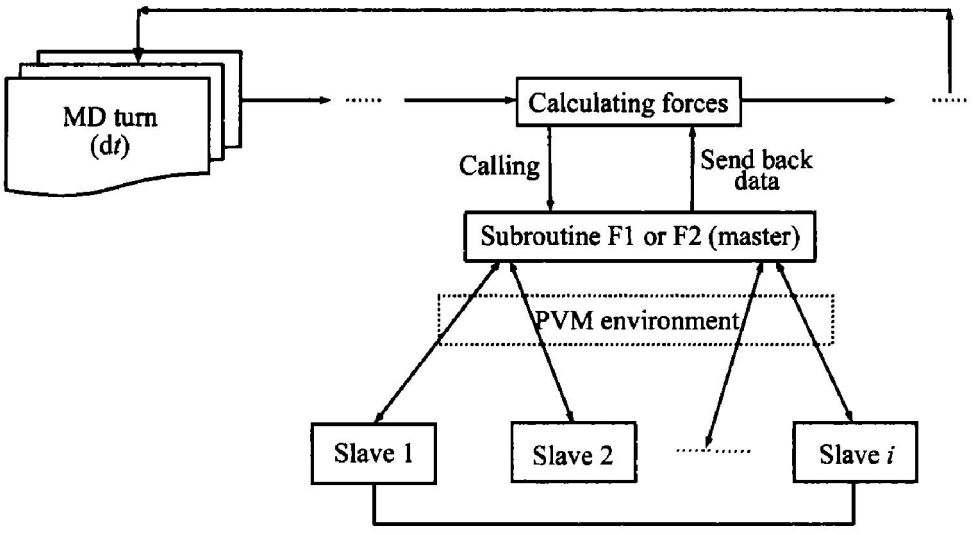

Fig. 1 Schematic diagram of parallel algorithm

original program, used to calculate the forces on each atom, have been reformed by parallel processing, and transformed into the slave programs. Calculating the forces, the master program will call the subroutine F1 and F2, and the subroutines will transfer the necessary data to each slave machine. As a slave gets the data, a circle calculation will be started. After finishing calculation, the slave will send the results to the master machine and then in the waiting state until the next turn begins. After the master machine has received all the results from the slaves in turn, the next turn will be continued until the whole simulation calculation ends up.

At present, we have simulated a system comprising 50 000 atoms, using 50 processing units of the YH-3M supper-computer for the parallel computing simultaneously. Adopting this parallel algorithm, the actual time consumption of each turn is closer to the 1/50 (about 600 s) of that we estimated by using cascade algorithm, and this time consumption is acceptable for us in computer simulation studies.

4 RESULTS AND ANALYSIS

We adopt the pair effective potential function of the generalized energy independent non-local model pseudo-potential theory developed by Wang et al[6,7] to simulate a system comprising 50 000 Al atoms:

$$V(r) = \frac{Z_{\text{eff}}^2}{r}\left[1 - \frac{2}{\pi}\int_0^\infty F(q)\,\frac{\sin(qr)}{q}\,\mathrm{d}q\right] \quad (4)$$

At 943 K (which is some higher than the melting point of Al, 933 K), we place 50 000 Al atoms in a cubic box, their initial coordinates are obtained randomly, and then let the whole system pre-run for 2 000 times to get an equivalent state at 943 K. Then the system is cooled to lower temperature at a rate of 4×10^{12} K/s. During the cooling process, for each temperature interval (in general about 50 K), the system state (include the coordinates, velocities, energies and so on of atoms) would be recorded, separately. And then the pair distribution function, the bond indexes and the clusters have been counted from the results at each temperature as follows.

4.1 Pair distribution function

The pair distribution function curve of the system is shown in Fig. 2. From Fig. 2, it can be seen that the simulation result of the larger system comprising 50 000 atoms is in good agreement with that from experiments. And there is a little difference between this simulation result and that from small-sized system comprising only 500 atoms. Since the pair distribution function $g(r)$ is a statistical average value,

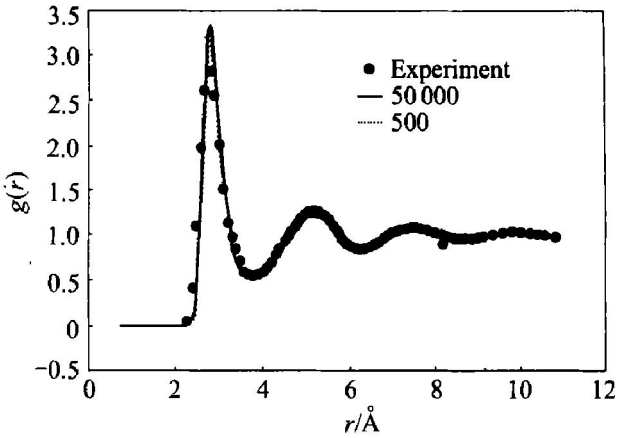

Fig. 2 Pair distribution function $g(r)$ of liquid metal Al at 943 K

some of the microscopic differences between two systems have been lost in averaging. From this we can see that if the purpose is to get the pair distribution function, it is enough to choose the small-sized system comprising 500 atoms. But it is not enough for getting important microstructure difference to utilize only this macroscopic statistical value.

4.2 Bond-type index analysis

We adopt the Honeycutt-Andersen (HA) bond-type index method[11], at present, which is an important method to study and analyze the microstructures of disorder systems such as liquid or amorphous metals. The changes of the percent relative numbers of various HA bond-types to total bonds with temperature during the cooling process is shown in Table 2.

Comparing these results with those obtained from the simulation of small-sized system comprising 500 atoms (as shown in Table 1 of Ref. [12]), we can see that though the changing tendency of bond-types of two systems are essentially the same, their concrete data still have some obvious differences. From this, it is just to be explained that the influence of the periodic boundary condition to the simulation results should not be neglected. For example, among the bond-types only the 1 551 bond-type increases remarkably. For the large-sized system, the relative number of 1 551 bond-type increases from 16.95% at 943 K to 29.256% at 300 K, while for the small-sized system the corresponding values are 16.6% and 26.0%, respectively (as shown in Table 1 of Ref. [12]). There is only a little difference of the relative number of bond-types at the initial state (943 K) of both systems, but at 300 K, their difference has amounted to 3.596%. From this, it can be clearly seen that the results obtained from the large-sized system comprising 50 000 atoms will closer to the actual situation than those

Table 2 Relationship of relative numbers of various bond-types with temperature(%)

Temperature/K	Bond-types									
	1 201	1 211	1 311	1 321	1 421	1 422	1 431	1 551	1 541	1 661
943	1.320	1.292	6.677	6.675	3.470	6.906	21.475	16.950	15.888	5.027
883	1.250	1.171	6.439	6.432	3.174	6.637	21.470	18.191	16.062	5.220
833	1.141	1.117	6.084	6.259	3.074	6.549	21.305	19.107	16.072	5.267
780	1.083	1.022	5.658	5.976	2.997	6.259	21.423	20.351	16.373	5.459
730	0.951	0.939	5.361	5.704	2.900	6.074	21.096	21.535	16.404	5.656
675	0.828	0.841	4.957	5.463	2.768	5.944	20.977	22.647	16.756	5.810
625	0.768	0.839	4.705	5.338	2.735	5.746	20.999	23.908	16.772	5.864
550	0.774	0.813	4.647	5.199	2.627	5.657	21.094	25.069	16.841	5.858
500	0.750	0.823	4.557	5.147	2.576	5.597	20.997	26.041	16.851	5.736
400	0.700	0.788	4.334	5.103	2.343	5.350	20.644	28.358	16.762	5.607
300	0.723	0.951	4.167	5.198	2.188	5.204	20.657	29.596	16.752	5.542
Tendency	– ①	–	–	–	–	–	–	+ + ③	+ ②	+

①– means decreasing; ②+ means increasing; ③+ + means increasing remarkably

obtained from the small-sized system, since the effects of the periodical boundary conditions above mentioned have been decreased remarkably.

4.3 Atomic clusters analysis

In liquid and amorphous metals, the direct reflection of short order is that there are a large number of cluster structures in the systems. We have also analyzed the clusters in the system comprising 50 000 atoms and obtained that of all the clusters the most remarkable in variation and the greatest in number is the icosahedron, as shown in Fig. 3.

From Fig. 3, we can see that the number of icosahedron increases nearly following a line. But there is an abnormal bend on the curve between 550 K and 450 K. According to the experiments, the glass transition temperature T_g is just in this temperature range. Therefore, it has been testified that the number of icosahedron has an obvious change at the phase transition point. As to the phase-transition process, the concrete situation of icosahedron structures should be studied in the future.

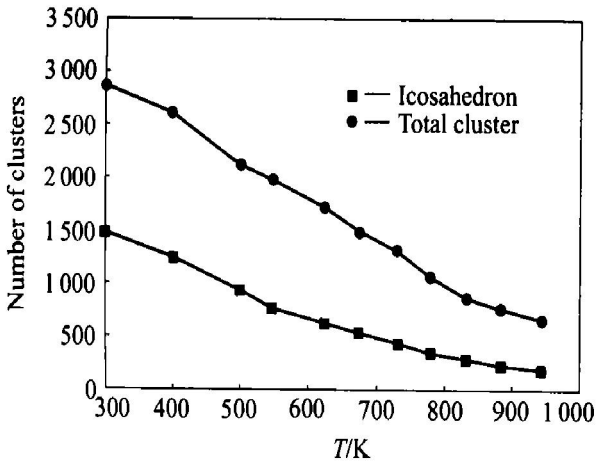

Fig. 3 Relationship of number of icosahedron with total number of all clusters

4.4 Visualizing analysis

1) The 2-Dimension section diagram of simulation system. Using computer graphics we can draw out the 2-dimension section diagram as shown in Fig. 4.

From Fig. 4, the atomic arrangements are random and long-range disorder. But in some parts we can see the obvious regularity, somewhere it ap-

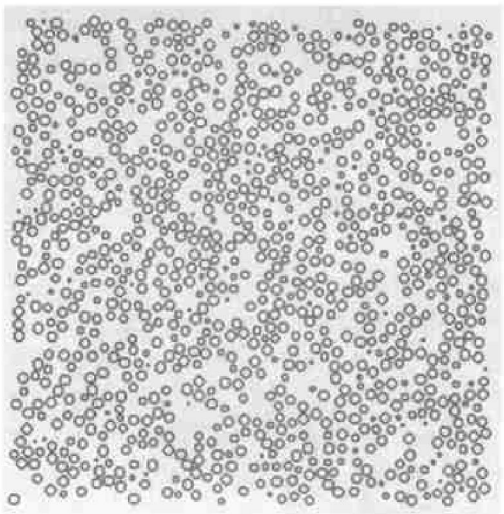

Fig. 4 2D section diagram plane (100)

pears to be the icosahedron structure, somewhere even to be the like-crystal arrangement, and somewhere there are obvious "empty holes". However, the whole appearance feature cannot be observed in the section diagram of the system comprising 500 atoms since their information are not enough.

2) Clusters in a system comprising 50 000 atoms. At present, the cluster structure is a problem to which there are more researches, since it is an important step to study the sub-microstructures of materials and their transition processes. In experiment, in common case the metal clusters are prepared by using special methods. But in the large-sized system of 50 000 atoms simulated here, we can find a great number of larger clusters that are composed of many clusters (mainly being icosahedron). However, these larger clusters are less found in the small-sized systems comprising only 500 atoms. Fig. 5 and Fig. 6 are the larger clusters that are composed of 3 icosahedra and 6 clusters (including the defect icosahedron), respectively. These results tell us that in the actual macroscopic materials, there are certainly some more complete and larger cluster structures, though they can not easily be observed in experiment at present.

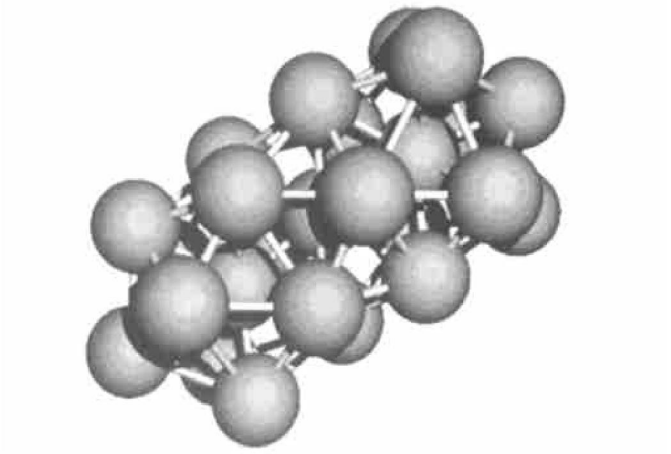

Fig. 5 Cluster comprising 3 icosahedra

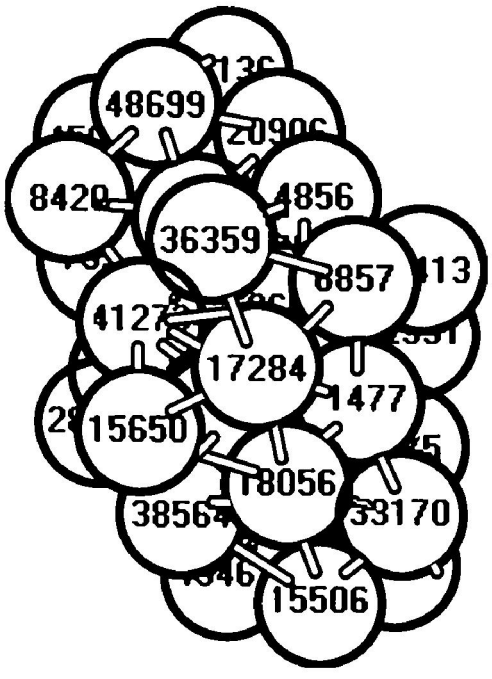

Fig. 6 Larger cluster comprising 6 clusters

5 CONCLUSIONS

Based on the deep analysis of the original MD simulation program, the parallel algorithm has been accomplished and it makes the number of atoms in simulating system amounted to 50 000 – 100 000. By this parallel algorithm, we have successfully performed the MD simulation studies on the large-sized systems comprising 50 000 Al atoms, and have got rather satisfied results more closed to the reality. We think that if the VERLET algorithm could be reconstructed entirely from the parallel algorithm, and adopting the higher performance super-computers, the total number of atoms in simulation system can be enlarged to tens, even hundreds times. Thus the simulation results will be more and more closed to the realities of materials.

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