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Prediction of the lowest energy configuration for Lennard-Jones clusters

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Based on the work of previous researchers, a new unbiased optimization algorithm—the dynamic lattice searching method with two-phase local search and interior operation (DLS-TPIO) —is proposed in this paper. This algorithm is applied to the optimization of Lennard-Jones (LJ) clusters with $N = 2$–650, 660, and 665–680. For each case, the putative global minimum reported in the Cambridge Cluster Database (CCD) is successfully found. Furthermore, for LJ_{533} and LJ_{536}, the potential energies obtained in this study are superior to the previous best results. In DLS-TPIO, a combination of the interior operation, two-phase local search method and dynamic lattice searching method is adopted. At the initial stage of the optimization, the interior operation reduces the energy of the cluster, and gradually makes the configuration ordered by moving some surface atoms with high potential energy to the interior of the cluster. Meanwhile, the two-phase local search method guides the search to the more promising region of the configuration space. In this way the success rate of the algorithm is significantly increased. At the final stage of the optimization, in order to decrease energy of the cluster further, the positions of surface atoms are further optimized by using the dynamic lattice searching method. In addition, a simple new method to identify the central atom of icosahedral configurations is also presented. DLS-TPIO has higher computing speed and success rates than some well-known unbiased optimization methods in the literature.

global optimization, Lennard-Jones clusters, interior operation, two-phase local search, dynamic lattice searching

1 Introduction

The cluster optimization problem, i.e., the problem of predicting lowest-energy structures of clusters, is one of the important topics in computational chemistry. Generally, the determination of lowest-energy structures of clusters can help us understand many chemical and physical properties of real clusters [1–3]. However, these problems are usually nondeterministic polynomial-time hard (NP-hard) due to the fact that the configuration space grows exponentially with the cluster size. Therefore, developing highly efficient global optimization methods plays an important role in tackling these problems both in theory and practice.

In the past 20 years, Lennard-Jones (LJ) clusters have been intensively investigated. In fact, they have become a benchmark system for evaluating the performance of global optimization algorithms. The LJ potential can be described as follows,

$$E_{\mathrm{LJ}} = 4\varepsilon \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^{6} \right] \quad (1)$$

where N denotes the size of the cluster, r_{ij} denotes the distance between atoms i and j, and the reduced units are used here, i.e., $\varepsilon = 1.0$, $\sigma = 1.0$. From eq. (1), it can be shown that the structural optimization of LJ clusters is essentially a continuous optimization problem.

A large number of methods have been employed to solve the problem, and representative examples include the basin-

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hopping method and its variants [4–7], simulated annealing algorithm (SA) [8, 9], hierarchical greedy algorithm (HGA) [10], genetic algorithms (GAs) [11–15], conformation space annealing algorithm (CSA) [16], adaptive immune optimization algorithm (AIOA) [17], random tunnel algorithm (RTA) [18], heuristic algorithm with surface and interior operators (HA-SIO) [19], the dynamic lattice searching algorithm (DLS) and its variants [20–23], the simple greedy method with energy-based perturbation as well as small population (EP-SGM) [24], and other algorithms [25, 26].

Methods for solving the problem can be divided into two categories, namely biased and unbiased methods [22]. In biased methods, a lattice, which is related to a special motif, is constructed in advance, such as an icosahedral (IC) lattice. Then the optimization is carried out based on the constructed lattice. This type of method is very fast, but the optimality of the results obtained cannot be guaranteed, due to the restriction of the search space. With this type of method, most of the putative global minima for LJ clusters up to $N = 1610$ can be located [15, 27–29]. On the other hand, unbiased methods start with a randomly generated local minimum and the optimization is performed based on random perturbations [22]. Using such methods, many putative global minima for LJ clusters were successfully found for the first time. For example, in 1997, the putative global minima of LJ_{69}, LJ_{78} and LJ_{107} were located by means of a basin-hopping method by Wales and Doye [4]. In 1999, Leary [7] located the global minimum of LJ_{98} with the MSBH (monotonic sequence basin-hopping) method. In 2006, Takeuchi [19] located the global minima of LJ_{506}, LJ_{521}, LJ_{537}, LJ_{538} and LJ_{541} by using the HA-SIO method. Compared with biased methods, the unbiased methods possess a larger search space, and hence their searching ability is also stronger.

The DLS method [20] and two-phase local search method [30] have been shown to be particularly efficient techniques for cluster optimization problems. The DLS method is very fast by virtue of properly utilizing structural information about the cluster under study, and it has been proved to be one of the most efficient methods for the optimization of atomic clusters. On the other hand, studies have indicated that the two-phase local search method can significantly improve the efficiency of global optimization algorithms by employing a properly modified potential function [31], and it has been successfully applied to a variety of optimization problems [30–32].

In order to further develop highly effective methods for the optimization of clusters, a novel unbiased algorithm—which we call the DLS-TPIO method—based on DLS, the two-phase local search method as well as a simple interior operation (presented in Section 2.1), is proposed in this paper. To evaluate the performance of the proposed method, it was applied to optimize LJ clusters with sizes $N = 2$–650, 660, and 665–680.

2 Computational method

The basic ideal of the proposed method is to combine the advantages of DLS, two-phase local search and interior operations. The efficacy of these methods in DLS-TPIO will be discussed in Section 3.

The DLS-TPIO method starts with a randomly generated local minimum. Then cycles of interior operation and two-phase local search are performed until the energy of the cluster is not improved during the last 10 attempts. The resulting configuration is further optimized by means of the DLS method. Furthermore, for large-size clusters (with a size no less than 500), an operation creating a central vacancy and a second DLS procedure are also carried out in the optimization. The DLS-TPIO algorithm is described in detail in the following sections.

2.1 Interior operation

In general, the energies of interior atoms are lower than those of surface atoms in a local-minimum structure of LJ clusters. On this basis, Takeuchi [19] proposed an interior operation in which some surface atoms with high energy are moved to the interior of the cluster in order to reduce their potential energies. In 2008, Shao and co-workers [22] presented a variant of this operation.

In this study, we present an interior operation which is similar to those previously proposed by other researchers. In the operation, the first m atoms with highest energy are moved into a sphere having a radius of $r = 0.5$, where m is an integer between 1 and 5, and the energy of atom i can be calculated according to eq. (2). The center of the sphere coincides with the center of mass of the cluster. The advantage of our interior operation is that it is easier to implement compared with previous examples.

$$E(i) = 4 \cdot \sum_{j \neq i}^{N} \left(\frac{1}{r_{ij}^{12}} - \frac{1}{r_{ij}^{6}} \right) \tag{2}$$

2.2 Two-phase local search

The two-phase local search method [30] involves two local searches. Firstly, a first local search is performed with a modified potential function $E_{\mathrm{LJ}}^{\mathrm{mod}}$ to guide the search to the more promising region of the configuration space. Then a second local search is performed with original potential function E_{LJ} starting from the result of the first local search.

The modified potential function used in this study can be written as follows:

$$E_{\mathrm{LJ}}^{\mathrm{mod}} = \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} \left[\frac{1}{r_{ij}^{10}} - \frac{1}{r_{ij}^{5}} + \mu \cdot r_{ij} + \beta \cdot \left(\max\left\{ 0,\ r_{ij}^{2} - (\eta \cdot R)^{2} \right\} \right)^{2} \right] \tag{3}$$

where $\mu = 0.1$, $\beta = 2.0$, $\eta \in \{1.0, 1.35\}$, $R = (N/17.77)^{1/3} + 1.0$ [22], r_{ij} denotes the distance between atoms i and j, and N is the size of the cluster.

We obtain eq. (3) by making a further modification of a modified potential function proposed by Locatelli and Schoen in ref. [30]. The difference between these two modified potential functions lies in the estimation of the diameter of the cluster. In eq. (3), the value is given with $\eta \cdot R$ and can be changed adaptively with N if η is fixed, since R depends only on N. However, in the modified potential function proposed by Locatelli and Schoen, the value is given in terms of a parameter D, the value of which must be determined empirically according to the cluster size. Therefore, eq. (3) is more convenient to employ in the optimization.

In eq. (3), $\beta \cdot (\max\{0, r_{ij}^2 - (\eta \cdot R)^2\})^2$ represents a penalization term of the cluster diameter, and $\mu \cdot r_{ij}$ is the penalization term of the distance between atoms i and j. It is clear that the modified potential function (3) favors spherical configurations, since large distances between atoms are penalized. Since spherical configurations are consistent with most global-minimum configurations of LJ clusters, the use of eq. (3) can efficiently guide the search and hence increase the success rate of the algorithm.

2.3 Dynamic lattice searching method

The dynamic lattice searching method was proposed by Shao, Cheng *et al*. in ref. [20]. The method starts with a randomly generated local minimum. Then, a cycle of dynamic lattice construction, dynamic lattice searching, and local search is performed until no new structure with a lower potential energy can be found. Dynamic lattice construction determines all the stable positions on the surface of the cluster for an added atom. These positions and N_{mov} (where N_{mov} is a predetermined integer) positions occupied by the atoms with highest energy are called dynamic lattice sites, and a set of these dynamic lattice sites is called a dynamic lattice. Dynamic lattice searching identifies several low-energy candidates by searching the dynamic lattice with a simple greedy method. A detailed description of the DLS method can be found in ref. [20].

The DLS method has been successfully applied to LJ clusters [20], Morse clusters [33], C_{60} molecular clusters [34], and Ag clusters [35]. The computational results have shown that the DLS method is highly efficient and unbiased.

2.4 Creation of a central vacancy

Because many global-minimum structures of large-size LJ clusters have IC configurations without a central atom [36], the central vacancy problem is also considered in this study. An operation creating a central vacancy is adopted in the DLS-TPIO method, where the central atom of the cluster is randomly moved on the surface of the cluster. In order to efficiently identify the central atom of a cluster, a definition—the atomic density—is introduced here. The atomic density of atom i can be defined as:

$$D(i) = \sum_{\substack{j=1(\neq i) \\ d_{ij} \leq \sqrt[6]{2}}}^{N} \frac{1}{d_{ij}^3} \tag{4}$$

where d_{ij} denotes the distance between atoms i and j, and N is the cluster size.

For those clusters with an IC configuration, the distances between atoms in the inner shell are usually smaller than those in the outer shell [37], since the interior atoms are compressed by the outer atoms in the cluster. Therefore, according to eq. (4), the atom with maximal atomic density is usually the central atom of a cluster.

2.5 Local searches

In the present study, the limited-memory quasi-Newton method (L-BFGS) [38] is adopted as a local search procedure. Previous studies have indicated that the L-BFGS method possesses strong local searching ability and usually outperforms the conjugate gradient method for in optimization of clusters [39]. In fact, it is already widely used in the structural optimization of clusters.

2.6 Description of the DLS-TPIO algorithm

(1) Randomly generate the initial structure of a cluster in a spherical container with a radius of $R = (N/17.77)^{1/3} + 1.0$, where the center of the spherical container coincides with the origin of the coordinates. Then the initial structure is locally optimized by means of the L-BFGS method, and the value of the parameter *NoImprove* is set to 0.

(2) Perform the interior operation and subsequent two-phase local search.

(3) If the energy of the cluster is improved, update the structure (i.e., replace the old structure by the current structure), reset the value of *NoImprove* to 0, and then return to step (2). Otherwise, maintain the former structure and increase the value of *NoImprove* by 1.

(4) If *NoImprove* < 10, then return to step (2). Otherwise, go to step (5).

(5) Carry out the DLS procedure with the current structure as the initial structure.

(6) If the energy of the current cluster is equal to or lower than the putative global minimum, or the size of the cluster is fewer than 500, then stop the calculation. Otherwise, go to the next step.

(7) Create a central vacancy, i.e., the central atom of the cluster is randomly moved on the surface of the cluster.

(8) Carry out the second DLS procedure with the current structure as the initial structure.

(9) Stop the calculation.

In the above algorithm, if the cluster size is no more than 309, then the number m of atoms moved by the interior operation takes a value of 1. Otherwise, the value of m is selected randomly from 1–5. In addition, for LJ_{38}, $LJ_{75–77}$, $LJ_{102–104}$, $LJ_{188–192}$, and $LJ_{236–238}$, which are considered as very difficult cases for most unbiased optimization methods in the literature, the value of η in eq. (3) is set to 1.0; for other cases, the value is set to 1.35. The above algorithm is summarized in Figure 1.

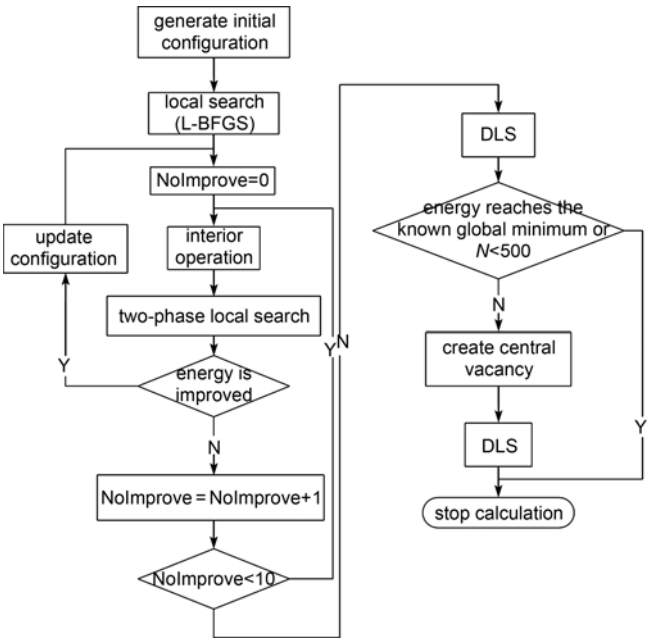

Figure 1 Flowchart of the DLS-TPIO algorithm.

3 Results and discussion

The DLS-TPIO algorithm was implemented in C language and all the calculations were carried out on a PC with 2.4 GHz CPU and 1 G RAM. The parameters used in DLS-TPIO are listed in Table 1, where the meaning of each parameter is the same as that in ref. [20]. On account of cost, only those clusters with sizes N = 2–650, 660, and 665–680 were optimized in this study. For each case in Tables 2–4 below, 250 independent runs of DLS-TPIO were performed.

By using DLS-TPIO to optimize the above-mentioned LJ clusters, all of the known global minima listed in the Cambridge Cluster Database (CCD) were successfully located (an updated list of the global minima of LJ clusters containing up to 1000 atoms can be found in the CCD: http://www-wales.ch.cam.ac.uk/CCD.html). Furthermore, for LJ_{533} and LJ_{536}, the potential energies obtained by the DLS-TPIO method are superior to the best previous results.

Table 1 Parameters used in the DLS-TPIO algorithm

N	N_{mov}	N_p	N_{best}	N_{try}
13–49	10	92	4	100
50–150	15	252	5	400
151–200	15	252	10	500
201–309	20	492	10	700
310–450	30	642	15	1000
451–561	40	812	15	1000
562–665	60	812	15	1500
666–680	80	812	20	1500

Table 2 Numbers of local searches needed by different methods for some selected cluster sizes

N	DLS-TPIO	HA-SIO[19]	DLS[20]	AIOA[17]	EP-SGM[24]	MSBH[6]	HGA[10]
38	56	2352	3240	3046	1739	2875	
50	96	51	270	537	264	460	
75	357	4374	2586	43000	86000	152000	9259
98	1138	3312	597	32000	27200	180000	5660
100	659	844	610	9304	2525	9128	
102	95	3881	7733	31000	13880	36028	
150	115	267	1398		3230		
188	1614	7320	12234				
200	124	2371	4494	140000	56550		
236	3299	16216	30875				
250	185	4611	11347				
300	112	2017	14875				
400	208	42600					
450	336						
500	695	52462	70000				62000
561	213	9419					

Table 3 Hit rates of DLS-TPIO for some selected cases

N	Hit rate	N	Hit rate
38 [b]	232/250	200 [a]	176/250
50 [a]	154/250	236 [b]	6/250
75 [b]	41/250	250 [a]	114/250
98 [b]	16/250	300 [a]	186/250
100 [a]	26/250	400 [a]	134/250
102 [b]	167/250	450 [a]	85/250
150 [a]	165/250	500 [a]	51/250
188 [b]	12/250	561 [a]	154/250

a) The optimal configuration is based on IC packing; b) the optimal configuration is based on non-IC packing.

The new and previous putative global minima of these two cases are listed in Table 5. Additionally, the new global-minimum structures for LJ_{533} and LJ_{536} have been deposited in the CCD.

In general, the average number of local searches needed to search for the global minimum structure is used as a criterion to evaluate the speed of unbiased optimization algorithms [6, 10, 17, 19–24]. A smaller number of local searches results in higher computing speed. The average number of local searches required by the DLS-TPIO method is listed for some selected cases in Table 2, and compared with those for some reference methods. It can be seen that the number of local searches needed by the DLS-TPIO method is much less than that for all of reference methods except for clusters with $N = 50$, 98, and 100. For example, for LJ_{75}, the numbers of local searches required by the HA-SIO [19] and DLS methods [20] are, respectively, 12 and 7 times larger than that for the DLS-TPIO method. For LJ_{500}, the numbers of local searches required by the HA-SIO [19] and HGA [10] methods are, respectively, 75 and 89 times larger than that for the DLS-TPIO method.

The variation in the number of local searches required to find the global minimum as a function of the cluster size for $N = 100$–580 is plotted in Figure 2. It can be seen that the value is less than 7×10^3 for each case in this size range. Furthermore, the value is less than 10^3 for most cases. Moreover, it also can be seen that peaks exist for $N = 188$, 236, 350, 486, and 578, which means that optimization of these cases is difficult with the DLS-TPIO method, as it is with other methods. On the other hand, comparison of Figure 2 with Figure 2 in ref. [19] shows that the number of local searches required using HA-SIO [19] is several to hundreds times larger than that for DLS-TPIO for most cases in the size range $N = 100$–561. These results show that the DLS-TPIO method is relatively fast.

The CPU time needed to find the global minimum is usually adopted as a criterion to assess the performance of global optimization algorithms [19–23]. The CPU time consumed by the DLS-TPIO method is given as a function of the cluster size for cluster sizes in the range $N = 100$–580 in Figure 3. It can be seen that the CPU time is less than 10^4

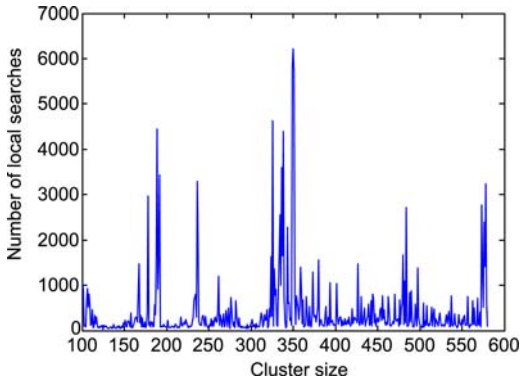

Figure 2 Number of local searches needed to find the global minimum.

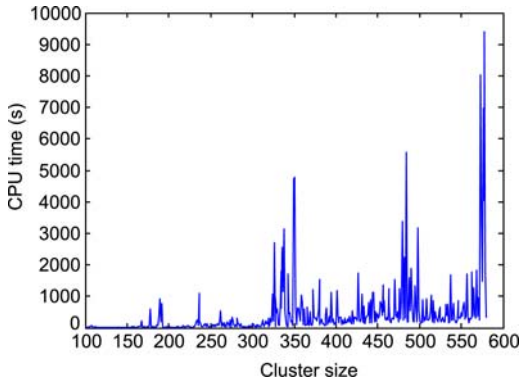

Figure 3 CPU time needed to find the global minimum.

seconds for all the cases and is less than 10^3 seconds for most cases. For LJ_N ($N = 100$, 200, 300, 400, 500, and 561), the average CPU times required to find the global minimum are 0.53, 0.467, 1.08, 3.95, 20.07, 8.78 min, respectively; however, using the HA-SIO method [19] (the calculations were performed on a computer with 3 GHz CPU and 1 G RAM), the corresponding values are 1.3, 16, 29, 1059, 2016, and 470 min, respectively.

The success rate is another important criterion to evaluate the performance of an algorithm [6, 17, 20–24]. To illustrate the efficiency of the proposed method, the success rates obtained in the optimization of some selected cases are given in Table 3. It can be seen that the success rate is very high for those cluster sizes where the optimal configuration is based on IC packing. For example, the value is 186/250 for LJ_{300}. The success rate is lower, but not unacceptably low, for those cluster sizes where the optimal configuration is based on non-IC packing. For example, for LJ_{98} and LJ_{188}, the values are 16/250 and 12/250, respectively. In addition, it should be noted that the DLS-TPIO method possesses very high success rates for LJ_{38} and LJ_{102}, which are very difficult to optimize using most unbiased optimization methods in the literature.

The dynamic lattice searching method with interior operation (DLS-IO) [22] is known to be one of most efficient unbiased methods for the optimization of the large-size

Table 4 Comparison of the results for the DLS-IO and DLS-TPIO methods

N	DLS-TPIO [b]			DLS-IO [b]		
	N_{LS} [a]	CPU time (h)	Hit rate (%)	N_{LS} [a]	CPU time (h)	Hit rate (%)
500	695	0.284	20.4	9943	10.76	0.6
561	213	0.146	60.2	9302	15.09	0.8
660	39745	43.3	0.4	124028	181.40	0.1
665	2649	2.893	6	152205	209.17	0.1
670	375	0.405	43.2	21208	43.02	0.6

a) N_{LS} represents the average number of local searches required for one hit of the global minimum; b) DLS-TPIO and DLS-IO were run on computers with 2.4 GHz CPU and 1 G RAM and with 2.8 GHz CPU, 1 G RAM, respectively.

Table 5 Comparison of the new and previous global minima

N	Energy [a]	Energy [b]
533	–3629.299922	–3628.252883 [c]
536	–3651.941851	–3651.779047 [c]

a) New global minima; b) previous global minima; c) previous global minima for LJ533 and LJ536 are taken from refs. [15] and [19], respectively.

clusters. To further illustrate the efficiency of the DLS-TPIO method, we compared the results of the DLS-TPIO method with those of the DLS-IO method. The results are shown in Table 4. It can be seen that the DLS-TPIO method is superior to the DLS-IO method in terms of the average number of local searches, CPU time, and success rate. For example, for LJ_{665}, the average number of local searches, CPU time, and success rate for the DLS-IO method are 152205, 209.17 h, and 0.1%, respectively; whereas the corresponding results for the DLS-TPIO method are 2649, 2.893 h, and 6%, respectively.

In the DLS-TPIO method, the interior operation, two-phase local search and DLS all play important roles. In the initial stage of the optimization, by moving some surface atoms with high energy to the interior of the cluster, the interior operation efficiently reduces the energy of the cluster and makes the configuration become gradually ordered [22]. Meanwhile, the two-phase local search guides the search to the more promising region of the configuration space. In this way the success rate of DLS-TPIO is greatly increased. At the final stage of the optimization, the positions of surface atoms are further optimized by means of the DLS method, and the energy of the cluster may be reduced again.

It should be noted that the DLS-TPIO method is still unbiased. Although the modified potential function in eq. (3) favors spherical configurations, other types of configurations also have a finite probability of being located due to the fact that the initial configuration of the cluster is generated randomly and the interior operation used in DLS-TPIO is stochastic. For example, we could obtain IC, decahedral (DE) as well as face-centered-cubic (FCC) configurations for LJ_{100} by using the DLS-TPIO method. Furthermore, the successful determination of lowest-energy structures with non-IC motifs also shows that DLS-TPIO is unbiased.

Additionally, computational results have indicated that the value of η in eq. (3) has a considerable influence on the formation of the configuration of a cluster. Taking LJ_{75} as a test case, the success rate obtained with the DLS-TPIO method is 41/250 when η is equal to 1.0; however, it is reduced to only 4/250 when η is equal to 1.35. A possible reason is that the compression forces on the interior atoms are different for different values of η, and the forces increase as the value of η decreases.

Finally, we should point out that although the DLS-TPIO method is very fast for most clusters in the size range $N \leqslant 680$, the optimization is still very time-consuming when the corresponding global-minimum configuration is based on DE packing. For example, for LJ_{660}, the CPU time required for searching the global minimum is as long as 43.3 h. Therefore, further improvements of the DLS-TPIO method are still needed.

4 Conclusion and future prospects

Based on the interior operation, two-phase local search and DLS method, a new unbiased optimization algorithm, called the DLS-TPIO algorithm, is proposed in this paper. LJ clusters in the size range $N = 2$–650, 660, and 665–680 were optimized using this algorithm, and all the global minima listed in the CCD were successfully located. Furthermore, for LJ_{533} and LJ_{536}, the energies obtained in this study are superior to the previous best results. Compared with some well-known unbiased methods in the literature, the DLS-TPIO algorithm has very high computing speeds and success rates. These results indicate that the DLS-TPIO algorithm is a powerful tool for the optimization of LJ clusters. In addition, the algorithm is also applicable to some other atomic clusters, such as Morse clusters.

We hope to apply the algorithm to other atomic clusters in the near future, and plan to adopt different diameter penalization functions in the two-phase local search method.

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