**Molecular dynamics investigation of structure evolution and thermodynamics of Ni-Fe nanoparticles during inert gas condensation**

Bei Lia,b,\*, Lei Pana, Changan Liua, Xu Zhanga

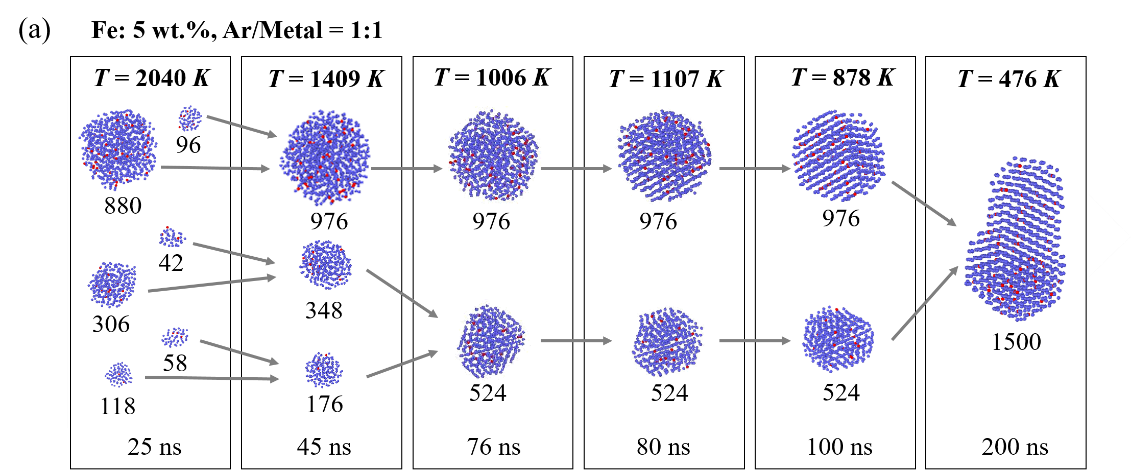
a School of Materials Science and Engineering, Research Center for Materials Genome Engineering, Wuhan University of Technology, Wuhan 430070, China

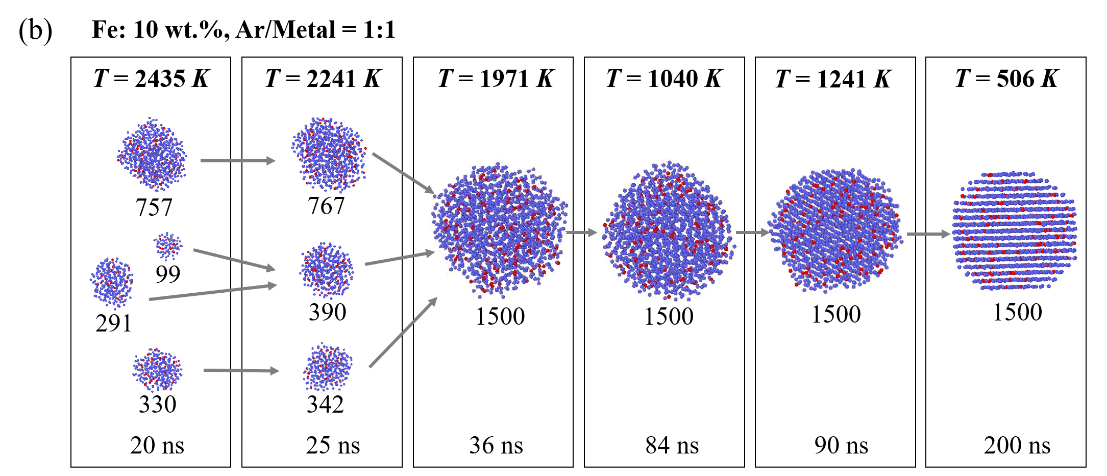
b State Key Laboratory of Materials Processing and Die & Mould Technology, Huazhong University of Science and Technology, Wuhan 430074, China

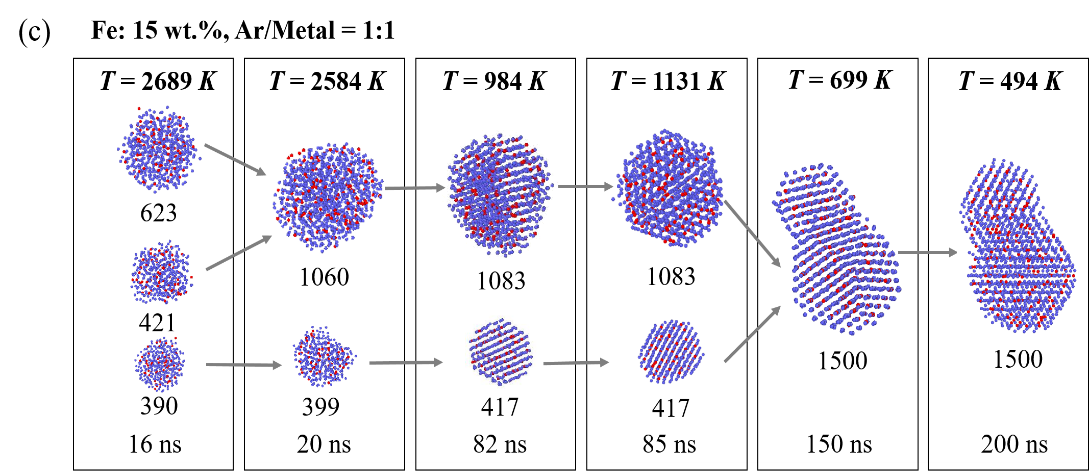
*\** Corresponding author. E-mail: libei@whut.edu.cn

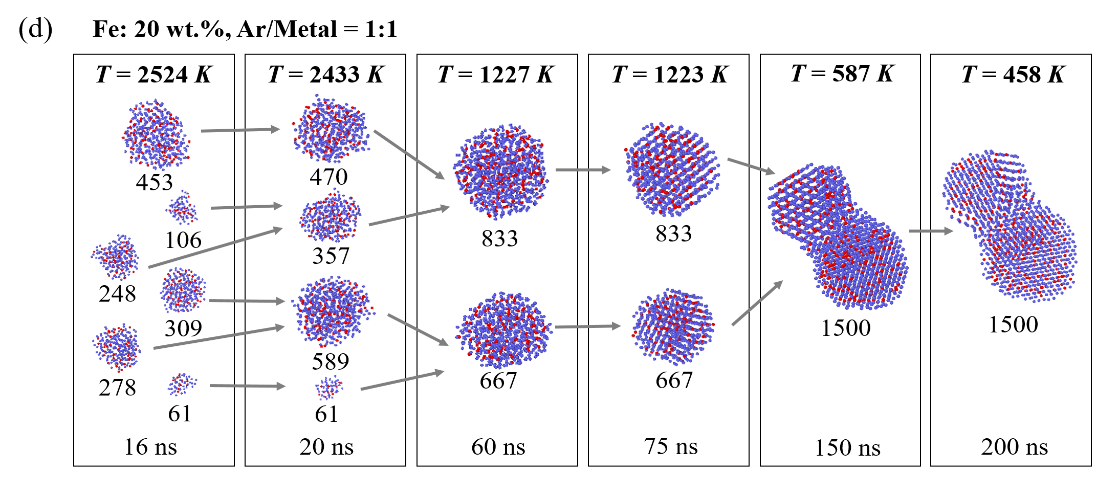
|  |  |
| --- | --- |
|  |  |
| (a) | (b) |
|  |  |
| (c) | (d) |

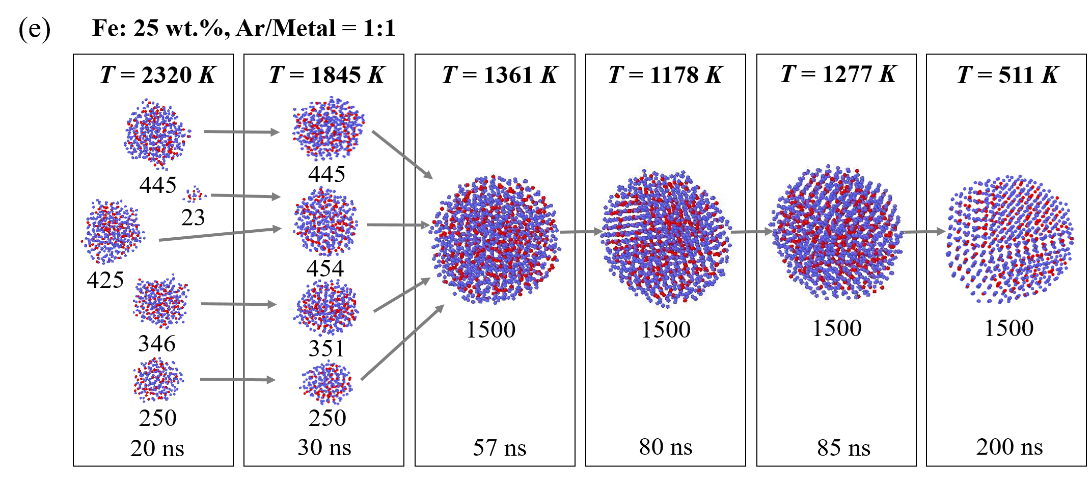
Fig. S1 Radial distribution function (RDF) profiles for different atomic pairs for Ni-Fe nanoparticles at 200 ns: (a) Fe: 10 wt.%, Ar/Metal = 1:1, (b) Fe: 30 wt.%, Ar/Metal = 1:1, (c) Fe: 30 wt.%, Ar/Metal = 0.75:1 and (d) Fe: 30 wt.%, Ar/Metal = 3:1.

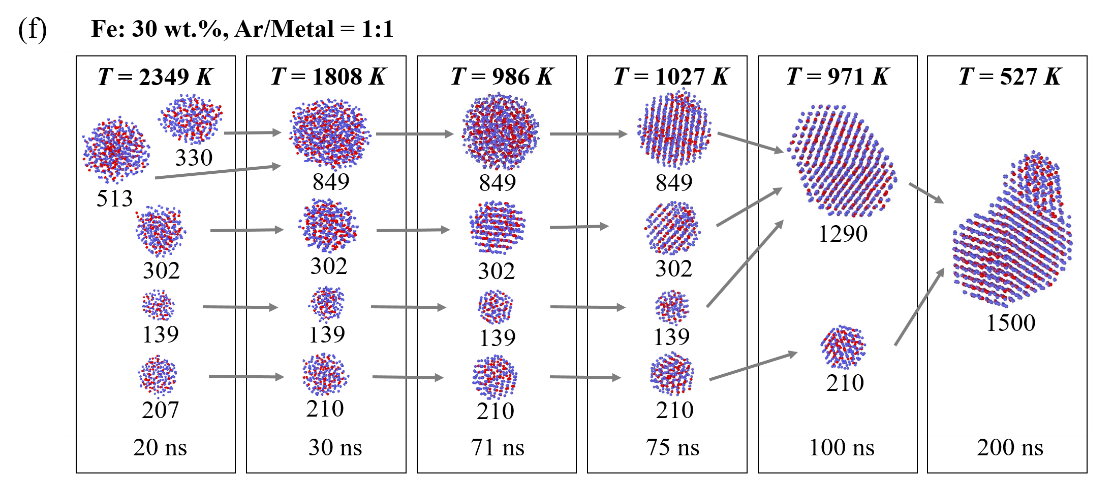


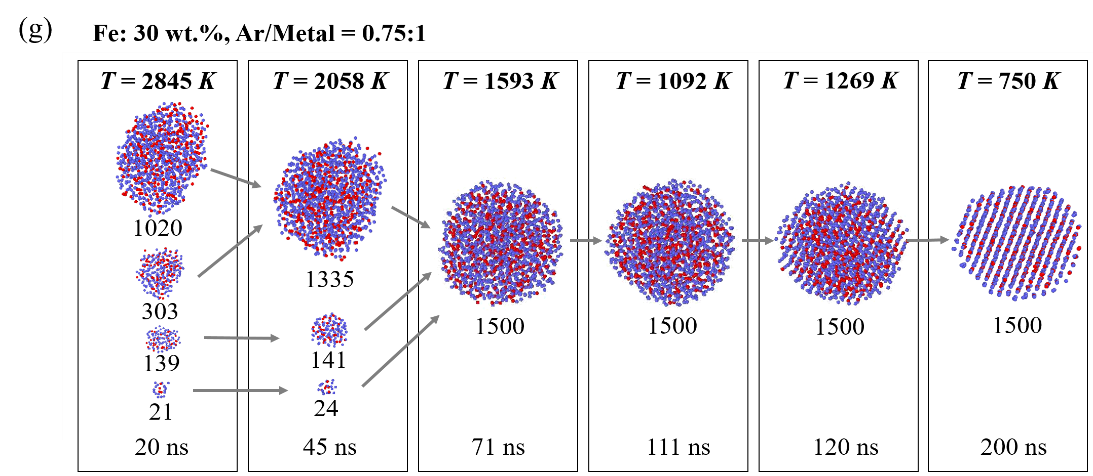


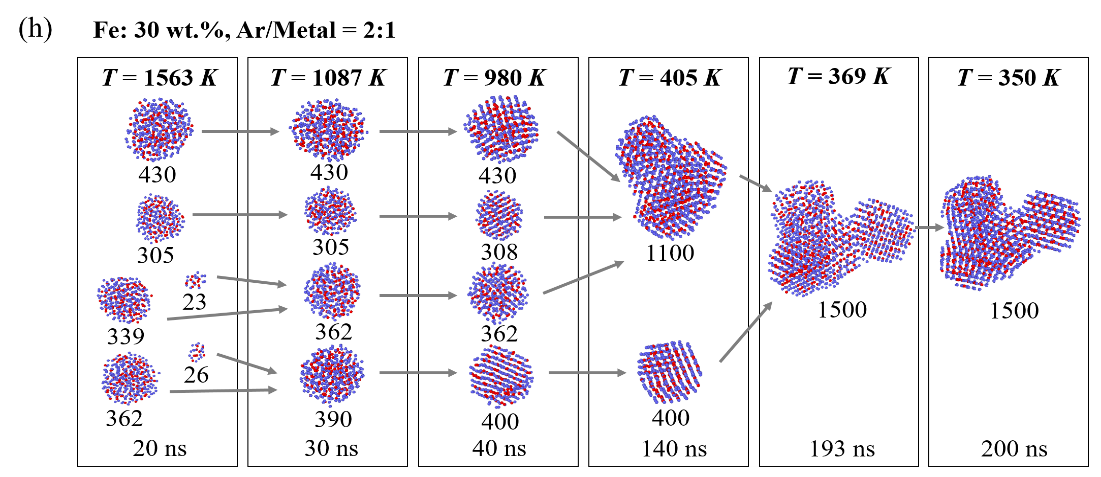


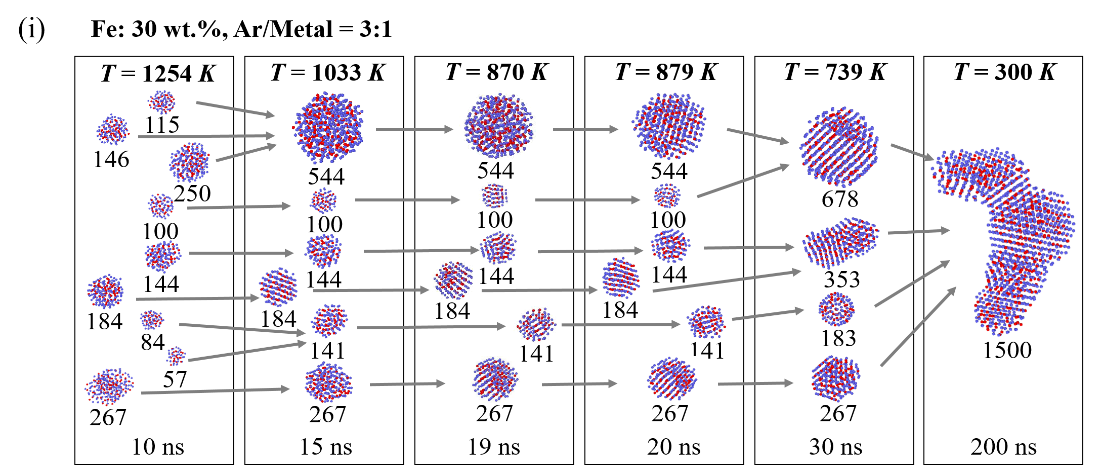












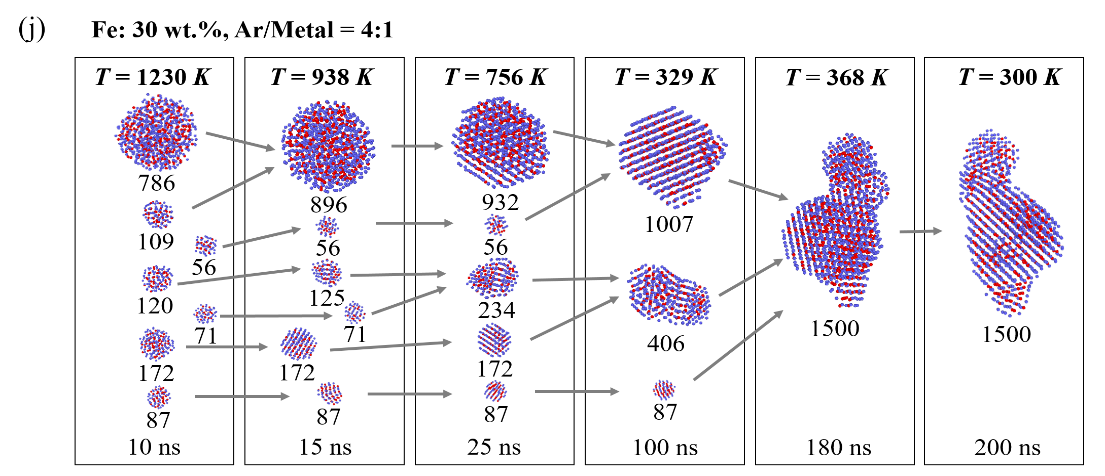
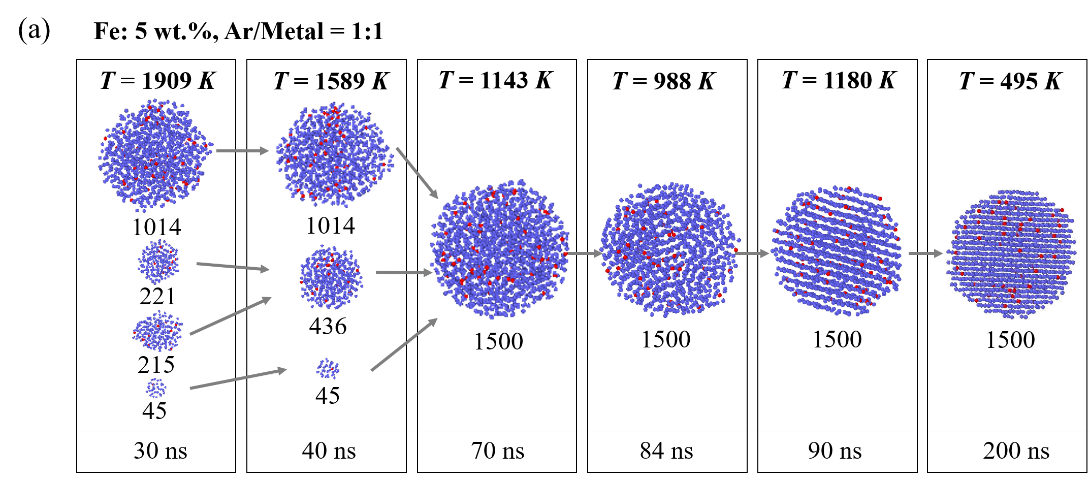


Fig. S2 Snapshots of the growth of Ni-Fe nanoparticles for (a) Fe: 5 wt.%, Ar/Metal = 1:1, (b) Fe: 10 wt.%, Ar/Metal = 1:1, (c) Fe: 15 wt.%, Ar/Metal = 1:1, (d) Fe: 20 wt.%, Ar/Metal = 1:1, (e) Fe: 25 wt.%, Ar/Metal = 1:1, (f) Fe: 30 wt.%, Ar/Metal = 1:1, (g) Fe: 30 wt.%, Ar/Metal = 0.75:1, (h) Fe: 30 wt.%, Ar/Metal = 2:1, (i) Fe: 30 wt.%, Ar/Metal = 3:1 and (j) Fe: 30 wt.%, Ar/Metal = 4:1 at different metal temperatures *T* and simulation time *t*. The number indicates the amount of metal atoms for each cluster. Fe and Ni atoms are colored red and purple, respectively.



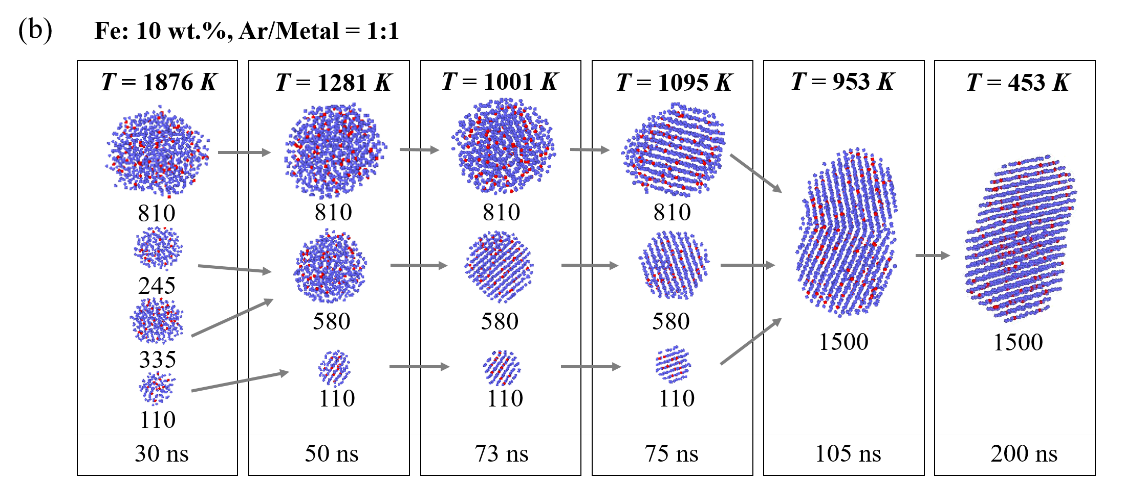


Fig. S3 Snapshots of the growth of Ni-Fe nanoparticles using different initial configurations for (a) Fe: 5 wt.%, Ar/Metal = 1:1 and (b) Fe: 10 wt.%, Ar/Metal = 1:1. Fe and Ni atoms are colored red and purple, respectively.

Table S1 Fe concentration in metal atoms, number of atoms (*N*s) and Fe concentration in the cluster *S* that solidifies at *t*s, and its corresponding solidification temperature *T*s obtained from MD simulations for different Fe concentrations.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Fe concentration  in metal atoms (wt.%) | *N*s | *t*s (ns) | Fe concentration  in the cluster *S* (wt.%) | *T*s (K) |
| 5 | 176 | 54.11 | 2.8 | 849.8 |
| 348 | 61.04 | 3.7 | 902.5 |
| 976 | 75.95 | 5.8 | 998.1 |
| 10 | 1500 | 83.67 | 10.0 | 1038.3 |
| 15 | 417 | 60.81 | 16.3 | 985.3 |
| 1083 | 81.55 | 14.5 | 1023.1 |
| 20 | 667 | 73.26 | 18.9 | 1013.1 |
| 833 | 72.68 | 20.9 | 1051.0 |
| 25 | 1500 | 78.66 | 25.0 | 1070.4 |
| 30 | 139 | 44.57 | 28.1 | 933.4 |
| 210 | 54.97 | 29.0 | 952.5 |
| 302 | 53.06 | 29.8 | 971.0 |
| 849 | 70.78 | 30.6 | 1055.9 |

Table S2 Ar/Metal ratio, number of atoms (*N*s) and Fe concentration in the cluster *S* that solidifies at *t*s, and its corresponding solidification temperature *T*s obtained from MD simulations for different amount of inert gas.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Ar/Metal ratio | *N*s | *t*s (ns) | Fe concentration  in the cluster *S* (wt.%) | *T*s (K) |
| 0.75:1 | 1500 | 110.77 | 30.0 | 1092.5 |
| 1:1 | 139 | 44.57 | 28.1 | 933.4 |
| 210 | 54.97 | 29.0 | 952.5 |
| 302 | 53.06 | 29.8 | 971.0 |
| 849 | 70.78 | 30.6 | 1055.9 |
| 2:1 | 305 | 30.95 | 30.8 | 989.1 |
| 362 | 33.82 | 27.6 | 967.1 |
| 390 | 33.40 | 31.0 | 1021.5 |
| 430 | 34.57 | 30.4 | 1004.8 |
| 3:1 | 100 | 11.28 | 28.0 | 833.5 |
| 141 | 15.42 | 33.3 | 885.1 |
| 144 | 14.10 | 27.1 | 919.1 |
| 184 | 13.48 | 31.0 | 954.0 |
| 267 | 16.43 | 31.5 | 963.7 |
| 544 | 18.87 | 29.2 | 1033.3 |
| 4:1 | 87 | 10.16 | 40.2 | 837.8 |
| 109 | 10.87 | 30.3 | 856.8 |
| 120 | 10.66 | 27.5 | 859.7 |
| 172 | 10.89 | 26.2 | 897.9 |
| 896 | 17.53 | 31.0 | 992.6 |