

Computational Analysis of SiC Crystal Growth from Silicon Melt Diluted with Cr, Fe, Co, Ni, Y, Al, La, Ce, Pr, Nd, and Sc. Part 1

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Abstract. The effect of various co-solvents on silicon carbide growth from solutions is sequentially analyzed within computational approach. The information related to the problem is collected from available literature and thoroughly treated. Boundary between liquid and solid state of solutions (liquidus line) is found from phase diagrams of 11 binary systems and is accounted for in calculating the carbon solubility at temperature and composition varying in a wide range. Thermophysical and transport properties are collected for preliminary estimation and comparison of growth rates. Their saturation with co-solvent percentage is predicted. Two-dimensional problem is set and first computations are demonstrated. It is shown that addition of lanthanum to the silicon melt gives a significantly higher growth rate than that of chromium.

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REFERENCES

- [1] G.Q. Liang, H. Qian, Y.L. Su, L. Shi, Q. Li, Y. Liu, Review of solution growth techniques for 4H-SiC single crystal, *China Foundry*, 2023, vol. 20, no. 2, pp. 159–178.
- [2] H.J. Scheel, *Introduction to liquid phase epitaxy*, in: P. Capper, M. Mauk (Eds.), *Liquid Phase Epitaxy of Electronic, Optical and Optoelectronic Materials*, John Wiley & Sons, 2007, pp. 1–19.
- [3] X.F. Liu, G.G. Yan, L. Sang, Y.X. Niu, Y.W. Heb, Z.W. Shen, Z.X. Wen, J. Chen, W.S. Zhao, L. Wang, M. Guan, F. Zhang, G.S. Sun, Y.P. Zeng, Defect appearance on 4H-SiC homoepitaxial layers via molten KOH etching, *J. Cryst. Growth*, 2020, vol. 531, art. no. 125359.

- [4] N. Komatsu, T. Mitani, T. Takahashi, K. Tomohisa, F. Kuniharu, T. Ujihara, Y. Matsumoto, K. Kurashige, H. Okumura, Growth rate and surface morphology of 4H-SiC single crystal grown under various supersaturations using Si-C solution, 2013, *Mater. Sci. Forum*, vol. 740–742, pp. 23–26.
- [5] H. Okamoto, Cr-Si (Chromium-Silicon), *Journal of Phase Equilibria*, 2001, vol. 22, no. 5, p. 593.
- [6] O. Kubaschewski, *IRON—Binary Phase Diagrams*, Springer Berlin, Heidelberg, 1982.
- [7] L. Zhang, Y. Du, H. Xu, Z. Pan, Experimental investigation and thermodynamic description of the Co–Si system, *Calphad*, 2006, vol. 30, no. 4, pp. 470–481.
- [8] E. Çadırlı, D.M. Herlach, T. Volkman, *J. Non-Cryst. Solids*, 2010, vol. 356, no. 9–10, pp. 461–466.
- [9] H. Okamoto, Si-Y (Silicon-Yttrium), *Journal of Phase Equilibria and Diffusion*, 2011, vol. 32, no. 5, pp. 475–476.
- [10] S. Ikhmayies, *Phase Diagrams of Al–Si System*, in: T. Wang, X. Chen, D.P. Guillen, L. Zhang, Z. Sun, C. Wang, N. Haque, J.A. Howarter, N.R. Neelameggham, S. Ikhmayies, Y.R. Smith, L. Tafaghodi, A. Pandey (Eds.), *Energy Technology 2019. The Minerals, Metals & Materials Series*, Springer, Cham, 2019, pp. 231–237.
- [11] H. Okamoto, La-Si (Lanthanum-Silicon), *Journal of Phase Equilibria and Diffusion*, 2007, vol. 28, no. 6, p. 585.
- [12] H. Okamoto. Ce-Si (Cerium-Silicon), *Journal of Phase Equilibria and Diffusion*, 2011, vol. 32, no. 5, pp. 470–471.
- [13] V.N. Eremenko, K.A. Meleshevich, Yu.I. Buyanov, Phazovaya diagramma sistemy Pr-Si, *Izvestiya Vuzov. Tsvetn. Metall.*, no. 3, pp. 82–87 (in Russian).
- [14] A.B. Gokhale, A. Munitz, G.J. Abbaschian, The Nd-Si (Neodymium-Silicon) system, *Bulletin of Alloy Phase Diagrams*, 1989, vol. 10, pp. 246–251.
- [15] H. Okamoto, Sc-Si (Scandium-Silicon), *Journal of Phase Equilibria*, 1992, vol. 13, no. 6, pp. 679–681.
- [16] D. Kondepudi, I. Prigogine, *Modern thermodynamics: from heat engines to dissipative structures*, John Wiley & Sons, Ltd, 2015.
- [17] T. Narumi, S. Kawanishi, T. Yoshikawa, K. Kusunoki, K. Kamei, H. Daikoku, H. Sakamoto, Thermodynamic evaluation of the C–Cr–Si, C–Ti–Si, and C–Fe–Si systems for rapid solution growth of SiC, *J. Cryst. Growth*, 2014, vol. 408, pp. 25–31.
- [18] L. Zhang, Y. Du, H. Xu, Z. Pan, Experimental investigation and thermodynamic description of the Co–Si system, *Calphad*, 2006, vol. 30, no. 4, pp. 470–481.
- [19] T. Tokunaga, K. Nishio, H. Ohtani, M. Hasebe, Thermodynamic assessment of the Ni–Si system by incorporating ab initio energetic calculations into the CALPHAD approach, *Calphad*, 2003, vol. 27, no. 2, pp. 161–168.
- [20] Z. Zhang, M.Y. Chu, X.S. Zhao, K. Li, S.L. Shang, Z.K. Liu, J.Y. Shen, Thermodynamic modeling of the Si-Y system aided by first-principles and phonon calculations, *Calphad*, 2019, vol. 65, pp. 282–290.
- [21] A. Qin, D. Liu, C. Chen, S. Liu, Y. Du, M. Wang, P. Nash, Heat contents of Sc₅Si₃ and ScSi intermetallics and thermodynamic modeling of the Sc–Si system, *Journal of Thermal Analysis and Calorimetry*, 2015, vol. 119, pp. 1315–1321.
- [22] H. Feufel, T. Gödecke, H.L. Lukas, F. Sommer, Investigation of the Al-Mg-Si system by experiments and thermodynamic calculations, *J. Alloys Compd.*, 1997, vol. 247, no. 1–2, pp. 31–42.
- [23] K. Xu, K. Chang, X. Zhou, L. Chen, J. Liu, Z. Deng, F. Huang, Q. Huang, Thermodynamic descriptions of the light rare-earth elements in silicon carbide ceramics, *J. Am. Ceram. Soc.*, 2020, vol. 103, no. 6, pp. 3812–3825.
- [24] L. Guo, G. Wang, J. Lin, Z. Guo, Z. Zhang, H. Li, Z. Cao, W. Yuan, Enhanced carbon solubility in solvent for SiC rapid solution growth: Thermodynamic evaluation of Cr–Ce–Si–C system, *J. Rare Earths*, 2023, vol. 41, no. 8, pp. 1272–1278.
- [25] A.F. Guillermet, Thermodynamic analysis of the Co–C System, *Int. J. Mater. Res.*, 1987, vol. 78, no. 10, pp. 700–709.
- [26] H. Ohtani, M. Yamano, M. Hasebe, Thermodynamic analysis of the Co–Al–C and Ni–Al–C systems by incorporating ab initio energetic calculations into the CALPHAD approach, *Calphad*, 2004, vol. 28, no. 2, pp. 177–190.

- [27] J. Gröbner, H.-L. Lukas, F. Aldinger, Thermodynamic calculations in the Y-Al-C system, *J. Alloys Compd.*, 1995, vol. 220, no. 1–2, pp. 8–14.
- [28] S. Kawanishi, K. Matsunaga, T. Yoshikawa, K. Morita, Thermodynamics and Kinetics of Direct Synthesis of Solar Grade Silicon from Metallurgical Silicon Wafer by Liquid Phase Migration in Solid Silicon, *Mater. Trans.*, 2017, vol. 58, no. 11, pp. 1571–1580.
- [29] Y. Peng, Y. Dua, L. Zhang, C. Sha, S. Liu, F. Zheng, D. Zhao, X. Yuan, L. Chen, Thermodynamic modeling of the C-RE (RE = La, Ce and Pr) systems, *Calphad*, 2011, vol. 35, no. 4, pp. 533–541.
- [30] Y. Bian, K. Tang, G. Tranell, A thermodynamic assessment of the Nd-C system, *Calphad*, 2015, vol. 51, pp. 206–210.
- [31] J. Kim, *Critical evaluation and thermodynamic modeling of Mg-RE-X (X = Mn, Si, Sn) systems coupled with experimental investigation*, Ph.D. Thesis, Department of Mining and Materials Engineering, McGill University, Montreal, QC, 2015.
- [32] T.Ya. Velikanova, L.V. Artyukh, S.M. Ilyenko, V.M. Danilenko, Phase equilibria in the Sc-M-C ternary systems, *Calphad*, 1998, vol. 22, no. 1, pp. 69–84.
- [33] K. Yanaba, M. Akasaka, M. Takeuchi, M. Watanabe, T. Narushima, Y. Iguchi, Solubility of carbon in liquid silicon equilibrated with silicon carbide, *Mater. Trans., JIM*, 1997, vol. 38, no. 11, pp. 990–994.
- [34] *Einstein relation (kinetic theory)*: *Wikipedia*, URL: [https://en.wikipedia.org/wiki/Einstein_relation_\(kinetic_theory\)](https://en.wikipedia.org/wiki/Einstein_relation_(kinetic_theory)) (last access: 02.04.2024).
- [35] E. Clementi, D.L. Raimondi, W.P. Reinhardt, Atomic Screening Constants from SCF Functions. II. Atoms with 37 to 86 Electrons, *J. Chem. Phys.*, 1967, vol. 47 no. 4, pp. 1300–1307.
- [36] L. Chybowski, M. Szczepanek, K. Gawdzińska, Arrhenius Equation for Calculating Viscosity in Assessing the Dilution Level of Lubricating Oil with Diesel Oil-A Case Study of SAE 30 and SAE 40 Grade Marine Lubricating Oils, *Energies*, 2024, vol. 17, no. 2, art. no. 444.
- [37] T. Iida, R.I.L. Guthrie, *The Physical Properties of Liquid Metals*, Clarendon Press, Oxford; Oxford University Press, New York, 1988.
- [38] M.J. Assael, K. Kakosimos, R.M. Banish, J. Brillo, I. Egry, R. Brooks, P.N. Quedstedt, K.C. Mills, A. Nagashima, Y. Sato, W.A. Wakeham, Reference Data for the Density and Viscosity of Liquid Aluminum and Liquid Iron, *J. Phys. Chem. Ref. Data*, 2006, vol. 35, no. 1, pp. 285–300.
- [39] F. Tesfaye Firdu, P. Taskinen, *Densities of Molten and Solid Alloys of (Fe, Cu, Ni, Co) - S at Elevated Temperatures - Literature Review and Analysis*, Aalto University Publications in Materials Science and Engineering, Espoo, 2010, pp. 1–33.
- [40] M.J. Assael, I.J. Armyra, J. Brillo, S.V. Stankus, J.Wu, W.A. Wakeham, Reference Data for the Density and Viscosity of Liquid Cadmium, Cobalt, Gallium, Indium, Mercury, Silicon, Thallium, and Zinc, *J. Phys. Chem. Ref. Data*, 2012, vol. 41, no. 3, art. no. 033101.
- [41] J.J. Valencia, P.N. Quedstedt, *Thermophysical Properties*, in: S. Viswanathan, D. Apelian, R.J. Donahue, B. DasGupta, M. Gywn, J.L. Jorstad, R.W. Monroe, M. Sahoo, T.E. Prucha, D. Twarog (Eds.), *ASM Handbook, Volume 15, Casting*, ASM International, 2010, pp. 468–481.
- [42] P.-F. Paradis, T. Ishikawa, N. Koike, Thermophysical Properties of Molten Yttrium Measured by Non-contact Techniques, *Microgravity Sci. Technol.*, 2009, vol. 21, pp. 113–118.
- [43] V.I. Kononenko, A.L. Sukhman, S.L. Gruverman, V.V. Torokin, Density and Surface Tension of Liquid Rare Earth Metals, Scandium, and Yttrium, *Phys. Stat. Sol. (a)*, 1984, vol. 84, no. 2, pp. 423–432.
- [44] C. Koyama, Y. Watanabe, Y. Nakata, T. Ishikawa, Density Measurement of Molten Scandium by an Electrostatic Levitator, *Int. J. Microgravity Sci. Appl.*, 2020, vol. 37, no. 3, art. no. 370303.
- [45] P.-F. Paradis, T. Ishikawa, N. Koike, Y. Watanabe, Study of Molten Lanthanum, Praseodymium, and Neodymium by Electrostatic Levitation, *J. Jap. Soc. Microgravity Appl.*, 2008, vol. 25, no. 3, pp. 407–412.
- [46] T. Ishikawa, J.T. Okada, J. Li, P.-F. Paradis, Y. Watanabe, *Thermophysical Properties of Liquid and Supercooled Rare Earth Elements Measured by an Electrostatic Levitator*, JAXA Research and Development Report, Japan Aerospace Exploration Agency, 2009, pp. 1–14.
- [47] J. Li, T. Ishikawa, J.T. Okada, Y. Watanabe, J. Yu, S. Yoda, Z. Yuan, Noncontact thermophysical property measurement of liquid cerium by electrostatic levitation, *J. Mater. Res.*, 2009, vol. 24, no. 7, pp. 2449–2452.

- [48] W.G. Rohr, The liquid densities of cerium and neodymium metals, *Journal of the Less Common Metals*, 1966, vol. 10, no. 6, pp. 389–391.
- [49] H. Daikoku, S. Kawanishi, T. Ishikawa, T. Yoshikawa, Density, surface tension, and viscosity of liquid Si–Cr alloys and influence on temperature and fluid flow during solution growth of SiC, *J. Chem. Thermodynamics*, 2021, vol. 160, art. no. 106476.
- [50] K.C. Mills, L. Courtney, Thermophysical Properties of Silicon, *ISIJ Int.*, 2000, vol. 40, pp. S130–S138.
- [51] R.K. Endo, Y. Fujihara, M. Susa, Calculation of the density and heat capacity of silicon by molecular dynamics simulation, *High Temp. High Press.*, 2003, vol. 35–36, no. 5, pp. 505–511.
- [52] Y. Sato, Representation of the Viscosity of Molten Alloy as a Function of the Composition and Temperature, *Jap. J. Appl. Phys.*, 2011, vol. 50, no. 11S, art. no. 11RD01.
- [53] T. Iida, R. Guthrie, M. Isac, N. Tripathi, Accurate Predictions for the Viscosities of Several Liquid Transition Metals, Plus Barium and Strontium, *Metall. Mater. Trans. B*, 2006, vol. 37B, pp. 403–412.
- [54] L. Battezzati, A.L. Greer, The viscosity of liquid metals and alloys, *Acta Metall.*, 1989, vol. 37, no. 7, pp. 1791–1802.
- [55] F.J. Cherne III, P.A. Deymier, Calculation of viscosity of liquid nickel by molecular dynamics methods, *Scr. Mater.*, 1998, vol. 39, no. 11, pp. 1613–1616.
- [56] Y. Sato, Y. Kameda, T. Nagasawa, T. Sakamoto, S. Moriguchi, T. Yamamura, Y. Waseda, Viscosity of molten silicon and the factors affecting measurement, *J. Cryst. Growth*, 2003, vol. 249, no. 3–4, pp. 404–415.
- [57] *STR Group: official website*, URL: <https://str-soft.com/> (last access: 02.04.2024).
- [58] *Thermophysical properties of materials for nuclear engineering: A tutorial and collection of data*, International Atomic Energy Agency, Nuclear Power Technology Development Section, Vienna, Austria, 2008.
- [59] M.J. Assael, A. Chatzimichailidis, K.D. Antoniadis, W.A. Wakeham, M.L. Huber H. Fukuyama, Reference correlations for the thermal conductivity of liquid copper, gallium, indium, iron, lead, nickel and tin, *High Temp. High Press.*, 2017, vol. 46, no. 6, pp. 391–416.
- [60] M.J. Assael, K.D. Antoniadis, W.A. Wakeham, M.L. Huber, H. Fukuyama, Reference correlations for the thermal conductivity of liquid bismuth, cobalt, germanium, and silicon, *J. Phys. Chem. Ref. Data*, 2017, vol. 46, no. 3, art. no. 033101.
- [61] *Yttrium properties*, URL: <https://material-properties.org/yttrium-thermal-properties-melting-point-thermal-conductivity-expansion/> (last access 02.04.2024).
- [62] *Scandium properties*, URL: <https://material-properties.org/scandium-thermal-properties-melting-point-thermal-conductivity-expansion/> (last access: 02.04.2024).
- [63] *Lanthanum properties*, URL: <https://material-properties.org/lanthanum-thermal-properties-melting-point-thermal-conductivity-expansion/> (last access: 02.04.2024).
- [64] I.V. Savchenko, D.A. Samoshkin, S.V. Stankus, Thermal Diffusivity Measurement of Cerium in the Temperature Range of 300–1800 K, *J. Eng. Thermophys.*, 2020, vol. 29, no. 1, pp. 42–48.
- [65] D.A. Samoshkin, A.Sh. Agazhanov, S.V. Stankus, Heat transfer coefficients of praseodymium in condensed state, *J. Phys.: Conf. Ser.*, 2019, vol. 1382, art. no. 012187.
- [66] D.A. Samoshkin, I.V. Savchenko, S.V. Stankus, A.Sh. Agazhanov, Thermal Diffusivity and Thermal Conductivity of Neodymium in the Temperature Range 293 to 1773 K, *J. Eng. Thermophys.*, 2018, vol. 27, no. 4, pp. 399–404.
- [67] J.B. Van Zytveld, Electrical resistivity of liquid chromium, *J. Non-Cryst. Solids*, 1984, vol. 61–62, part 2, pp. 1085–1090.
- [68] *Technical data for the element Iron in the Periodic Table*, URL: <https://periodictable.com/Elements/026/data.html> (last access: 02.04.2024).
- [69] *Technical data for the element Cobalt in the Periodic Table*: URL: <https://periodictable.com/Elements/027/data.html> (last access: 02.04.2024).
- [70] *Technical data for the element Yttrium in the Periodic Table*, URL: <https://periodictable.com/Elements/039/data.html> (last access: 02.04.2024).
- [71] *Technical data for the element Scandium in the Periodic Table*, URL: <https://periodictable.com/Elements/021/data.html> (last access: 02.04.2024).
- [72] G. Krieg, R.B. Genter, A.V. Grosse, Electrical conductivity of liquid lanthanum, *Inorg. Nucl. Chem. Lett.*, 1969, vol. 5, no. 10, pp. 819–823.

- [73] V.G. Postovalov, E.P. Romanov, V.P. Kondrat'ev, Structural Characteristics and the Temperature Derivative of the Electrical Resistivity of Liquid Lanthanides, *Phys. Metals Metallogr.*, 2007, vol. 103, no. 3, pp. 234–245.
- [74] *Technical data for the element Praseodymium in the Periodic Table*, URL: <https://periodictable.com/Elements/059/data.html> (last access: 02.04.2024).
- [75] *Technical data for the element Neodymium in the Periodic Table*, URL: <https://periodictable.com/Elements/060/data.html> (last access: 02.04.2024).
- [76] I. Barin, *Thermochemical Data of Pure Substances*, VCH, Weinheim, Federal Republic of Germany, 1995.
- [77] M.J. Blandamer, J.C.R. Reis, *A Notebook for Topics in Thermodynamics of Solutions and Liquid Mixtures*, LibreTexts, 2024, p. 1.15.5: Heat Capacities: Isochoric: Liquid Mixtures: Ideal (last access: 02.04.2024).
- [78] R.L. Rowley, G.L. White, M. Chiu, Ternary liquid mixture thermal conductivities, *Chem. Eng. Sci.*, 1988, vol. 43, no. 2, pp. 361–371.
- [79] C.Y. Ho, M.W. Ackerman, K.Y. Wu, T.N. Havill, R.H. Bogaard, R.A. Matula, S.G. Oh, H.M. James, Electrical Resistivity of Ten Selected Binary Alloy Systems, *J. Phys. Chem. Ref. Data*, 1983, vol. 12, no.2, pp. 183–322.
- [80] J.-Y. Yoon, M.-H. Lee, Y. Kim, W.-S. Seo, Y.-G. Shul, W.-J. Lee, S.-M. Jeong, Enhancement in the rate of the top seeded solution growth of SiC crystals via a roughening of the graphite surface, *Jap. J. Appl. Phys.*, 2017, vol. 56, no. 6, art. no. 065501.
- [81] T. Mitani, N. Komatsu, T. Takahashi, T. Kato, K. Fujii, T. Ujihara, Y. Matsumoto, K. Kurashige, H. Okumura, Growth rate and surface morphology of 4H-SiC crystals grown from Si-Cr-C and Si-Cr-Al-C solutions under various temperature gradient conditions, *J. Cryst. Growth*, 2014, vol. 401, pp. 681–685.