**Supporting Information for**

**Thermodynamic criteria of the End-of-Life silicon wafers refining for closing the recycling loop of photovoltaic panels**

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**Number of pages: 36**

**Figures: 2**

**Tables: 10**

**References: 140**

# **S1.　Metallurgical refining processes of metallurgical-grade silicon (MG-Si)**

The conventional production and purification of solar grade silicon (SoG-Si) starts from carbothermic reduction of high-purity quartz (SiO2) above 1800 ºC using coke to produce the metallurgical-grade silicon (MG-Si) of approximately 98% purity. MG-Si is then purified to produce electronic-grade silicon (EG-Si) of extremely high purity (>11N) using a chemical gaseous purification technique known as the Siemens method.[1] Conventionally, the raw materials for SoG-Si are from off-grade products during the purification process of EG-Si. However, the supply of off-grade EG-Si materials for SoG-Si is facing an increasing risk owing to the improvement of the EG-Si fabrication technology and the promotion of the scrap recycling inside the EG-Si process scope. Further, the Siemens purification process is extremely complicated and energy intensive. With such concerns, lots of effort have been made to promote the SoG-Si supply from alternative resources.

In past decades, large efforts have been made to produce SoG-Si by direct refining MG-Si. [2–9] Particularly, the high-temperature refining process including (1) solvent refining [4,10] with different solvents such as aluminum [11,12], copper [13], iron [14–16], nickel [17], and other metals [18–20]; (2) oxidative refining using various slags [21–24]; (3) vacuum refining by electron beam melting, plasma melting and so on [25–27]; (4) directional solidification [28]; and (5) electro-refining [29,30] have been extensively focused on.

# **S2. The activity coefficients of impurity elements in molten silicon**

The Redlich-Kister-type polynomial is always employed to illustrate the excess Gibbs energy for solution mixing in the CALPHAD approach[31]. The interaction parameters for the binary system of Si-M system described by Redlich-Kister polynomial have been assessed in lots of literature studies, and the activity coefficients of the impurity elements can be calculated using the assessed interaction parameters. The activity coefficient of impurity element M in the binary Si-M system can be calculated by the following polynomials derived from Redlich-Kister polynomial.

|  |  |
| --- | --- |
|  | (S1) |

Since the concentration of impurity elements in the silicon is extremely low in most cases, using activity coefficients of M in the dilute solution,****, is more correct. **** can be calculated much simpler at the limiting case of****:

|  |  |
| --- | --- |
|  | (S2) |

where(*p*=0, 1, 2, 3) are the temperature-dependent binary interaction parameters and expressed as follows:

|  |  |
| --- | --- |
|  | (S3) |

where *p*A, *p*B, *p*C, and *p*Dare constants

The calculated activity coefficients of impurity elements in silicon at 1773 K are listed in **Table S1**. The temperature-dependent binary interaction parameters,  (*p*=0, 1, 2, 3), for the assessment of the liquid phase in Si-M binary system obtained in the literatures are shown in **Table S2**.

**Table S1.** The activity coefficient of impurity elements in infinite dilute liquid silicon solution at 1773 K in pure liquid standard state

|  |  |  |  |
| --- | --- | --- | --- |
| M | *γ*M(l) | M | *γ*M(l) |
| Ag | 3.28 | Mo | 2.35E-03 |
| Al | 0.50 | Na | 2.00E-02 |
| Au | 8.37E-02 | Nb | 1.46E-04 |
| B | 3.87 | Ni | 2.90E-03 |
| Be | 5.29 | Os | 5.08E-03 |
| Bi | 30.49 | P | 0.37 |
| C | 2.64 | Pb | 36.38 |
| Ca | 2.35E-03 | Pd | 1.32E-03 |
| Ce | 9.67E-10 | Pt | 1.54E-05 |
| Co | 7.16E-03 | Re | 5.12E-04 |
| Cr | 1.16E-02 | Ru | 0.15 |
| Cu | 0.32 | Sb | 5.09 |
| Fe | 2.55E-02 | Sn | 7.49 |
| Ga | 1.52 | Ta | 1.25E-03 |
| Gd | 7.33E-06 | Ti | 2.29E-04 |
| Ge | 1.50 | U | 2.16E-04 |
| Hf | 1.43E-05 | V | 4.73E-03 |
| In | 4.57 | W | 7.81E-03 |
| La | 7.95E-08 | Y | 2.12E-07 |
| Mg | 0.364 | Zn | 1.48 |
| Mn | 8.978E-03 | Zr | 2.67E-05 |

.

**Table S2.** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the liquid phase in Si-M binary systems.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| M |  |  |  |  |  | Ref. |
| Ag | 11052.0-0.09508T | 16066.4-5.31210T |  |  |  | Chevalier 1988 [32] |
| Al | -11340.1-1.23394T | 3530.9-1.35993T | 2265.4 |  |  | Gröbner 1996 [33] |
| Au | -24103.30-15.14T | 29375.28-1.11T | -13032.24 |  |  | Meng 2007 [34] |
| B | 2400+9.89T |  |  |  |  | Yoshikawa 2005 [35] |
| Be | -28151.67+27.75T | -4949.18 | 8455.14 |  |  | Pan 2005 [36] |
| Bi | 46370+2.26T |  |  |  |  | Olesinski 1985 [37] |
| C | 25645-6.38T |  |  |  |  | Gröbner 1996 [33] |
| Ca | -228147.9+61.7892T | 110482.3-73.61283T | 44663.2-6.95691T | -82284+56.03509T |  | Anglezio 1994 [38] |
| Ce | -99974.63-61.61T | 63423-90.35T |  |  |  | Gröbner 2004 [39] |
| Co | -183483.8+34.80023T | 3219.5+15.28341T | 34241.7 | -15579.7 |  | Zhang 2006 [40] |
| Cr | -128000+21.23883T | 50016.61-14.31913T |  |  |  | Chen 2009 [41] |
| Cu | -38763.5+12T | 52431.2-27.4571T | -29426.5+14.775T |  |  | Yan 2000 [42] |
| Fe | -164435+41.977T | 21.523T | 5220+5.726T | 28955-26.275T |  | Miettinen 1999[43] |
| Ga | 14900-4.9T |  |  |  |  | Olesinski 1985 [44] |
| Gd | -265000+60T | -60000+25T |  |  |  | Huang 2007 [45] |
| Ge | 6610-0.354T |  |  |  |  | Jung 2010 [46] |
| Hf | -177631+6.43T | 1830 |  |  |  | Zhao 2000 [47] |
| In | 45100-12.8T |  |  |  |  | Olesinski 1985 [48] |
| La | -249912.5-39.8T | -25123.5 | 83012.5 | 21695.6 |  | Zhou 2010 [49] |
| Mg | -73623.6+20.9297T | 40644.7-23.7571T | -37653.7+35.7315T | -80905.8+44.1741T | -12484.9 | Lüdecke 1986 [50] |
| Mn | -152854+32.125T | 38217+4.414 | 42606-16.74T | -32556 |  | Lee 2012 [51] |
| Mo | -158013.345+12T | -20000+15.015T | 39026.3+5.1567T | 2461.9-5.5087T |  | Guo 2012 [52] |
| Na | -58420+16.94T | -47005+9.99T |  |  |  | Hao 2012 [53] |
| Nb | -199000 | 18800 | 50000 |  |  | Geng 2009 [54] |
| Ni | -205000+33T | 102700-27T | 25000 | -117000+55T |  | Miettinen 2005 [55] |
| Os | -125865+31.56T | -27569+11.07T |  |  |  | Liu 2001[56] |
| P\* |  | | | | | Yuan 2017 [57] |
| Pb | 66630-7.70T |  |  |  |  | Olesinski 1984 [58] |
| Pd | -253536.9+56.50T | 238372.9-86.77T | -127556.8+64.18T | -174993.5+71.85T | 178995.5-82.65T | Du 2006 [59] |
| Pt | -285834.40+32.71T | 110230.07-16.34T | 99818.56-27.74T | -111425.53+24.83T |  | Xu 2008 [60] |
| Re | -96700 | 45000 | -60000 |  |  | Shao 2001 [61] |
| Ru | -127858-27.62T | -59628+23.85T | 50985 |  |  | Liu 2001 [56] |
| Sb | 15463.82+4.81T |  |  |  |  | Wang 2011 [62] |
| Sn | 41468.97-10.94T | -30861.57+21.70T |  |  |  | Long 2012 [63] |
| Ta | -234317.343+44.55T | 77520.5861-11.68T |  |  |  | Drouelle 2013 [64] |
| Ti | -255852.17+21.874T | 25025.35-2.0023T | 83940.65-6.71526T |  |  | Seifert 1996 [65] |
| U | -185537+26.42 | -98478+53.79 | 47133-16.79T |  |  | Berche 2009 [66] |
| V | -190326.8+44.06T | -6265.4 | 39546.5 |  |  | Zhang 2008 [67] |
| W | -140000+35.1T | -20897.8 | -14660.85 |  |  | Li 2013 [68] |
| Y | -262885.3+87.63T | 5429.8-1.81T | -76199.8-25.40T |  |  | Ran 1989 [69] |
| Zn | 9692.3-1.29635T | -7943.3+3.57871T |  |  |  | Mey 1986 [70] |
| Zr | -200000+15.2454T | 10T |  |  |  | Chen 2009 [71] |

\* The activity coefficient of phosphorus in infinite dilute silicon melts was evaluated and described by the shown equation.

# **S3. The activity coefficients of impurity elements in solid silicon**

The temperature-dependent binary interaction parameters for the assessment of the silicon-rich solid solution phase (diamond phase) in Si-M binary system obtained in the literatures are shown in **Table S3**. The activity coefficient of the considered impurity elements in the dilute solid silicon can be calculated using the similar method as **Equation S1** shown above.

**Table S3** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the silicon-rich solid solution phase in Si-M binary systems.

|  |  |  |  |
| --- | --- | --- | --- |
| M | \* | | Ref. |
| A | B |
| Ag | 280000 | -71.5 | Yoshikawa, et al. 2010 [72] |
| Al | 93200 | -14.5 | Yoshikawa, et al. 2010 [72] |
| B | 80800 | -26.3 | Yoshikawa, et al. 2010 [72] |
| Bi | 251000 | -74.7 | Yoshikawa, et al. 2010 [72] |
| Ca | 80 |  | Anglezio, 1994 [38]  Yoshikawa, et al. 2010 [72] |
| Cu | 162000 | -39.6 | Yoshikawa, et al. 2010 [72] |
| Fe | 137650 | -8.1 | Tang and Tangstad, 2012 [73] |
| Ga | 67700 | -3.81 | Yoshikawa, et al. 2010 [72] |
| In | 207000 | -53.8 | Yoshikawa, et al. 2010 [72] |
| P | -40498.5 | 21.8079 | Liang and Fetzer, 2014. (model I) [74] |
| Sb | 67795.81 |  | Wang, et al. 2011 [62] |
| Sn | 18725.62 | 22.2896 | Long, et al. 2012 [63] |
| Ti | 133000 | -41.2 | Yoshikawa, et al. 2010 [72] |

\* 

# **S4. The activity coefficients of impurity elements in molten solvent metals: aluminum, copper, iron, lead, tin, and zinc.**

The calculated activity coefficients of the impurity elements in different solvent metals and the used literature interaction parameters for the liquid Sol.-M (Sol.=Al, Cu, Fe, Pb, Sn, Zn) systems are listed in **Table S4**, while the activity coefficients were set as unity as the first approximation for some systems where the assessment results were unavailable. The temperature-dependent binary interaction parameters for the assessment of the liquid phase in Sol.-M (Sol.=Al, Cu, Fe, Pb, Sn, Zn) binary system obtained in the literatures are shown in **Table S5 − Table S10**. The activity coefficient of the considered impurity elements in the liquid solvent metals can be calculated using the similar method as **Equation S1** shown above.

**Table S4.** The activity coefficients of impurity elements in different solvent metals.

|  |  |  |  |
| --- | --- | --- | --- |
| M | *γ*M(l) in Aluminium  (905 K) | *γ*M(l) in copper  (1200 K) | *γ*M(l) in iron  (1600 K) |
| Ag | 0.011 | 3.15 | 860.8 |
| Al | 1.00 | 0.00030 | 0.012 |
| B | 5.31 | 3.70 | 0.017 |
| Bi | 1.01 | 4.37 | 82.26 |
| Ca | 0.00025 | 0.014 | 8421 |
| Cu | 0.011 | 1.00 | 13.94 |
| Fe | 0.00011 | 58.05 | 1.00 |
| Ga | 1.10 | 0.00075 | 1.00 |
| In | 22.22 | 0.042 | 36.82 |
| P | 0.0014 | 0.000082 | 0.0000032 |
| Sb | 0.41 | 0.011 | 1.75 |
| Sn | 7.42 | 0.0099 | 1.73 |
| Ti | 0.00042 | 0.48 | 0.029 |
|  |  |  |  |
| M | *γ*M(l) in lead  (800 K) | *γ*M(l) in tin  (800 K) | *γ*M(l) in zinc  (800 K) |
| Ag | 3.63 | 0.59 | 0.75 |
| Al | 177.30 | 4.82 | 8.09 |
| B | 1.00 | 1.66×1011 | 30.14 |
| Bi | 1.00 | 1.37 | 19.24 |
| Ca | 8.74×10-8 | 1.85×10-8 | 5.56×10-8 |
| Cu | 1.00 | 0.24 | 0.026 |
| Fe | 180950.88 | 98.97 | 3.64×106 |
| Ga | 14.31 | 1.56 | 1,76 |
| In | 1.00 | 0.87 | 5.47 |
| P | 1.00 | 1.96 | 1.20×10-19 |
| Sb | 0.75 | 0.41 | 1.03 |
| Sn | 2.16 | 1.00 | 3.72 |
| Ti | 1.00 | 0.00021 | 0.087 |

**Table S5.** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the liquid phase in the Al-M binary systems.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| M |  |  |  |  | Ref. |
| Ag | -15300-8.8T | -25400+4.3T | 910+0.2T | 8500 | Lim 1995 [75] |
| B | -927.53-5.11T | 31961.6 | -12992 |  | Mirkovic 2004 [76] |
| Bi | 75.35-0.053T | 20.42-0.0037T |  |  | Kim 2006[77] |
| Ca | -89545+26.37T | -21847+11.77T | 4780+5.03T |  | Ozturk 2005 [78] |
| Cu | -67094+8.56T | 32148-7.12T | 5915-5.89T | -7290+5.5T | Liang 2015 [79] |
| Fe | -91976.5+22.13T | -5672.6+4.87T | 121.9 |  | Jacobs 2009 [80] |
| Ga | 2613.3-2.95T | 692.4-0.092T | 319.5 |  | Watson 1992 [81] |
| In | -10267+85.65T-0.055T2 | 8787-12.02T+0.0066T2 |  |  | Kaban 2010 [82] |
| P | -51769.9 |  |  |  | Liang 2013 [83] |
| Sb | -13328-5.10T | 10748+0.34T |  |  | Balakumar 2005 [84] |
| Sn | 16329.85-4.98T | 4111.97-1.15T | 1765.43-0.57T |  | COST 1998 [85] |
| Ti | -118048+41.97T | -23613+19.70T | 34757-13.84T |  | Witusiewicz 2008[86] |

**Table S6.** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the liquid phase in the Cu-M binary systems.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| M |  |  |  |  |  | Ref. |
| Ag | 16914.95-14.77T+1.55TlnT | -1963.30+0.86T |  |  |  | He 2006 [87] |
| Al | -67094+8.56T | -32148+7.12T | 5915-5.89T | 7290-5.5T |  | Liang 2015 [79] |
| B | -3156.2 | -14390.26+9.98T | 42620-20T |  |  | Wang 2009 [88] |
| Bi | 23844.75-9.84T | 1260.32+1.19T |  |  |  | Teppo, 1990 [89] |
| Ca | -27967 | -9738 | -4994 |  |  | Risold 1996 [90] |
| Fe | 35625.8-2.19T | -1529.8+1.15T | 12714.4-5.19T | 1177.1 |  | Chen 1995 [91] |
| Ga | -58110.5+154.54T-18.38TlnT | -22884.7+1.92T | -11256.9 |  |  | Li 2008 [92] |
| In | -41564.8+238.62T-19.83TlnT | -76057.8+371.31T-44.99TlnT | -42076.5+192.40T-23.23TlnT |  |  | Liu 2002 [93] |
| P | -180379+101.07T | 39496-91.51T | 35583 |  |  | Noda 2009 [94] |
| Sb | -16154.82+24.00T-4.03TlnT | -35130.8+50.33T-5.23TlnT | -29263.28+15.32T | -2300.89 | 8873.94 | Liu 2000 [95] |
| Sn | -9935.17-5.16T | -21571.21+4.84T | -11005.75-2.61T |  |  | Li 2013 [96] |
| Ti | -19330+7.65T |  | 9382-5.45T |  |  | Kumar 1996[97] |

**Table S7.** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the liquid phase in the Fe-M binary systems.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| M |  |  |  |  | Ref. |
| Ag | 81089 | 8806 |  |  | Swartzendruber 1984 [98] |
| Al | -91976.5+22.13T | 5672.6-4.87T | 121.9 |  | Jacobs 2009 [80] |
| B | -126220+29.44T | -8390 | 33538 |  | Rompaey 2002 [99] |
| Bi | 62327.74-4.39T | 3362.48 |  |  | Boa 2006 [100] |
| Ca | 120233 |  |  |  | Anglezio 1994 [38] |
| Cu | 35625.8-2.19T | 1529.8-1.15T | 12714.4-5.19T | -1177.1 | Chen 1995 [91] |
| In | 52838.5-5.54T | 3999.9 |  |  | Ohno 2009 [101] |
| P | -266000+41.3T | 96900-40.84T |  |  | Ohtani 2006 [102] |
| Sb | -20094.45+16.48T | -17411.15+11.60T |  |  | Boa 2006 [100] |
| Sn | 108744.39-441.227T+52.18TlnT | -8992.66+2.03T | -9855.25+0.98T | 2552.59 | Huang 2010 [103] |
| Ti | -74300+17.84T | 8299.85-6.10T |  |  | Bo 2012 [104] |

**Table S8.** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the liquid phase in the Pb-M binary systems.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| M |  |  |  | Ref. |
| Ag | 13330.32-6.21T | 1449.03+1.06T | -2089.13 | Lee 1994 [105] |
| Al | 47933.60-10.72T | -14407.33+6.65T | 4742.36-0.72T | Yu 1996 [106] |
| Ca | -104625.76-4.35T |  |  | Idbenali 2008 [107] |
| Fe | 110114.85-9.11T | 27699.55-6.74T |  | Vaajamo 2011 [108] |
| Ga | 13658.15+2.96T | -1420.89+1.39T | 5214.18-4.04T | Mathon 1996 [109] |
| Sb | -212.66-2.00T | -88.23 |  | Lee 1994 [105] |
| Sn | 6200-0.42T | 790-1.91T |  | Ohtani 1995 [110] |

**Table S9.** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the liquid phase in the Sn-M binary systems.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| M |  |  |  |  | Ref. |
| Ag | -399.49-31.42T+3.08TlnT | 18150.65-5.88T | -12009.03+5.18T |  | Gierlotka 2012 [111] |
| Al | 16329.85-4.98T | -4111.97+1.15T | 1765.43-0.57T |  | COST 507 1998 [85] |
| B |  | | | | Ma 2012 [112] |
| Bi | 446.6+0.88T | 1.21T |  |  | Lee 1996 [113] |
| Ca | -110358+3.18T | -10605 |  |  | Ohno 2006 [114] |
| Cu | -21571.21+4.84T | 11005.75+2.61T |  |  | Li 2013 [96] |
| Fe | 108744.39-441.227T+52.18TlnT | 8992.66-2.03T | -9855.25+0.98T | -2552.59 | Huang, 2010 [103] |
| Ga | 3369.7+0.039T | -528.9+0.11T |  |  | Anderson 1992[115] |
| In | -711-1.69T | 64+1.36T |  |  | Lee 1996 [113] |
| P | 16690-16.90T | 9696 | -8400 |  | Miettinen 2001 [116] |
| Sb | -5695.1-1.71T | -782.6 | 1840.9 |  | ManasijevićŽ 2008 [117] |
| Ti | -91598.90-0.94T | 45682.64-12.10T |  |  | Yin 2007 [118] |

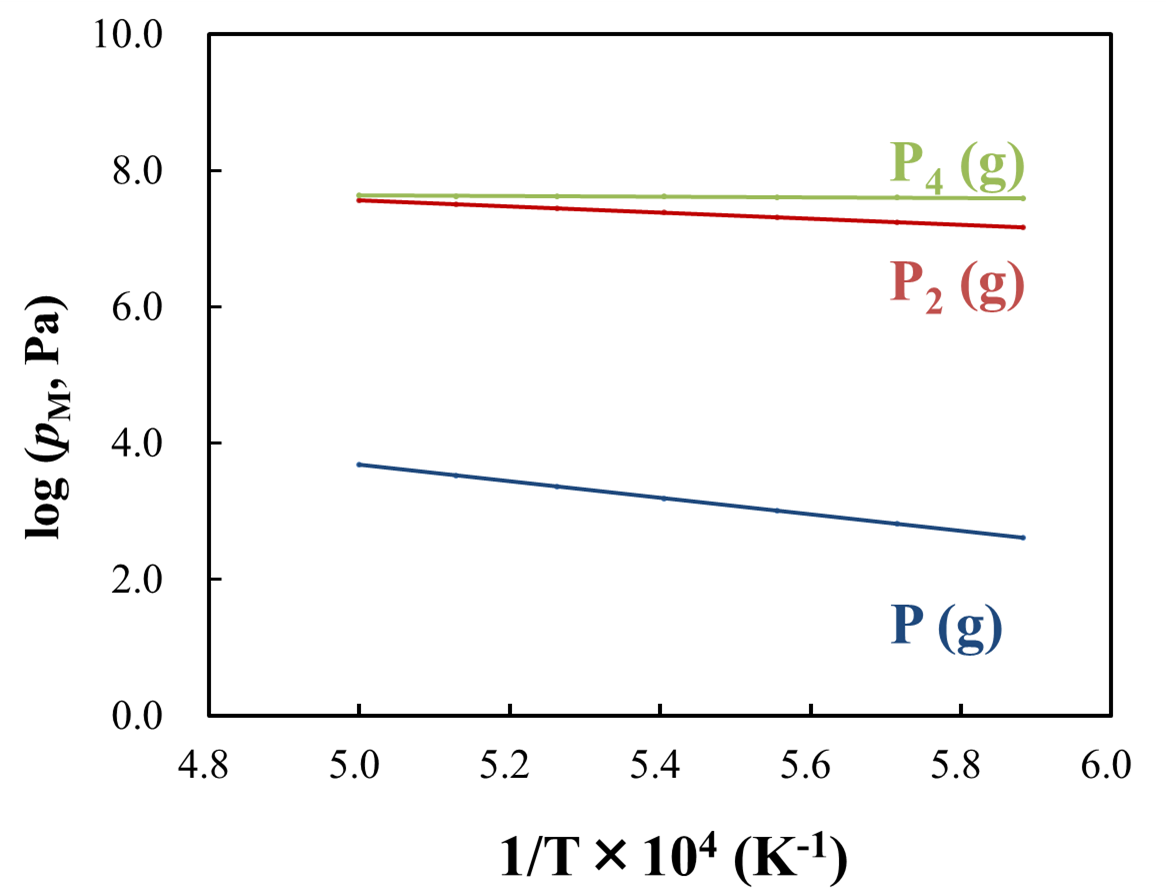
**Table S10.** The temperature-dependent binary interaction parameters (Redlich-Kister parameters) for the assessment of the liquid phase in the Zn-M binary systems.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| M |  |  |  |  | Ref. |
| Ag | -27400+5.88 | 5500 |  |  | Gomez-Acebo 1998 [119] |
| Al | 10288+3.04T | 810+0.47T |  |  | Chen 1993 [120] |
| B | 91314.2-66.68T |  |  |  | Chen 2013 [121] |
| Bi | 17731-8.08T+0.044TlnT | 1988+3.50T+0.16TlnT | 3104+0.066T-0.16TlnT | 10934+11.30T+0.29TlnT | Djaballah 2005 [122] |
| Ca | -65435+10.62T | -43288+12.98T | -49709+25.55T |  | Brubaker 2001 [123] |
| Cu | -89687+369.49T-44.58TlnT | 4756-2.20T | 10513-6.78T |  | Wang 2011 [124] |
| Fe | 58088-23.67T | 92219-55.58T | 13570 |  | Nakano 2005 [125] |
| Ga | 3898.25+26.09T-4.09TlnT | -874.29 |  |  | Mathon 2000 [126] |
| In | 12401-4.45T | 3186-1.78T | 679 |  | Lee 1996 [127] |
| P | -264052.22exp(0.0001447T) | 13108.03exp(-0.00084T) |  |  | Liu 2016 [128] |
| Sb | -11951-1.12T | 3325.6-1.00T | 29432-23.63T |  | Li 2007 [129] |
| Sn | 12558-8.70T | 5623-4.20T | 4149-4.09T |  | Lee 1996 [127] |
| Ti | -30000+17.2T |  |  |  | Doi 2006 [130] |

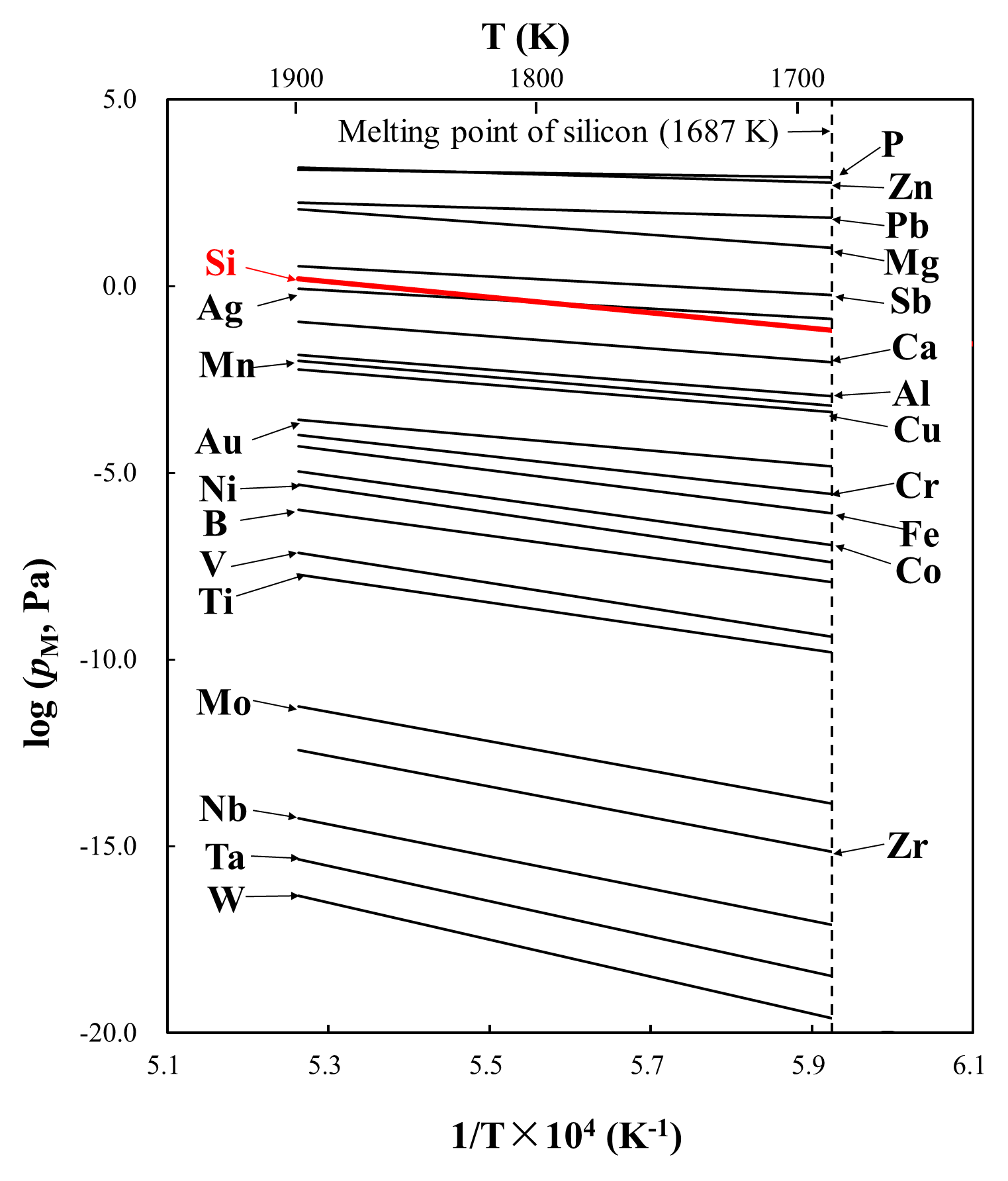
# **S****5.** **Effect of the temperature on evaporation refining**

Efficiency of the evaporation refining is determined by the difference of vapor pressures of impurity elements and that of molten silicon. Comparison of the equilibrium vapor pressures of phosphorus gas in forms of P (g), P2(g), and P4 (g) over pure phosphorus was shown in **Figure S1**. It was noticed that the dominant vapor of phosphorus is P4 (g) under the equilibrium condition at the considered temperature range.

**Figure S2** systematically compares the vapor pressures of 23 impurity elements in the dilute concentration and that of the pure molten silicon at different temperatures. The dependency of the elimination of the impurity elements by evaporation refining on temperature does not change significantly, though the vapor pressure of the impurity element generally increases as the temperature increase. Some of the impurity elements, including phosphorus, zinc, lead, magnesium, and antimony, have higher vapor pressures, even in dilute concentration than that of pure silicon. Evaporation refining is suitable to eliminate these impurity elements. Meanwhile, a lower temperature is found to have slight advantages in the elimination of these impurity elements. Since the vapor pressure of silver is approximately the same as that of pure silicon, elimination of silver by evaporation refining can only work when some loss of silicon through evaporation is acceptable. For other impurity elements, however, evaporation refining process is not suitable even at high temperatures. It is especially difficult to eliminate boron the typical dopants in PV cells, using the evaporation refining process.



**Figure S1** Comparison of the equilibrium vapor pressures of phosphorus gas in forms of P (g), P2(g), and P4 (g) over pure phosphorus.

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**Figure S2** Comparison of the vapor pressures of impurity elements in dilute concentration in silicon melt and that of pure molten silicon.

# **REFFERENCE**

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