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Optimal thermal plasma processes for zirconium carbide powder production from zircon concentrates

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Abstract

Available results of equilibrium composition and total enthalpy computation for the systems: Zr–Si–O–Ar and Zr–Si–O–C–H–Ar, presented in the paper, enables thermodynamic optimization of the plasma process for zirconium carbide production from zircon. In the first step of the considered process, zircon decomposition to zirconia and silica is followed by their evaporation in argon thermal plasma. In the second step by introduction of methane or propane–butane gas in the Zr–Si–O–Ar system and reactive quenching, the temperature of such obtained Zr–Si–O–C–H–Ar is lowered to a range in which it is possible to deposit zirconium carbide powder. On presented examples the procedure of determination of optimal mass, energy and temperature process parameters is demonstrated. From presented data, it is possible to calculate and optimize process parameters for any other thermodynamic conditions. © 2001 Elsevier Science Ltd and Techna S.r.l. All rights reserved.

Keywords: Zirconium carbide; Thermal plasma; Ceramic powder; Processing

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1. Introduction

Zirconium carbide (ZrC_x) has typical properties of transition metal carbides: elevated hardness — between alumina and diamond — a high melting temperature of 3420°C (from some sources 3540°C) and high chemical stability at room temperature [1,2]. ZrC has been used in nuclear plants as a low neutron absorbing and high mechanical resistant refractory material and also as a constituent in cathodes made of UC–ZrC [3]. It is also used in the form of tubes, boats and crucibles to handle molten metals and as part of electronic devices used for thermoionic transducers. Recently ZrC has been used along with zirconium diboride, ZrB_2, to produce composite materials [4,5].

Different methods have been used to produce ZrC; one method is by direct reaction of zirconium hydride or halide with carbon. Another method used is direct reduction and carburization of zirconium oxide, ZrO_2(s). The experimental work to investigate the possibility of ZrC powder production directly from zircon concentrates by reacting it with methane in a thermal plasma reactor [6], was performed in the Department of Metallurgical Engineering at the College of Mines and Earth Resources, University of Idaho (USA). A mixture of argon–nitrogen gas was used as plasma gas. The main constituents found in collected powder samples were zirconium oxide, zirconium carbide, silicon carbide and some silica. The existence of unreacted zirconia indicates that not all of it was reduced, probably due to the fact that it did not vaporize completely [6]. For this reason we have decided to choose thermodynamic consideration of the thermal plasma process for zirconium carbide production from zircon concentrates in argon plasma. Due to a greater temperature difference in argon thermal plasma there are better conditions (than in plasma of a nitrogen–argon mixture) for complete evaporation of zirconia before reacting with methane, propane–butane or any other reducing gas.

Before the paper was finished, by kindness of Prof. Patrick R. Taylor (University of Idaho) we received Ref. [7], from which we have seen that experiment of the plasma process for zirconium carbide production from zircon concentrates, was much more successfully realized.

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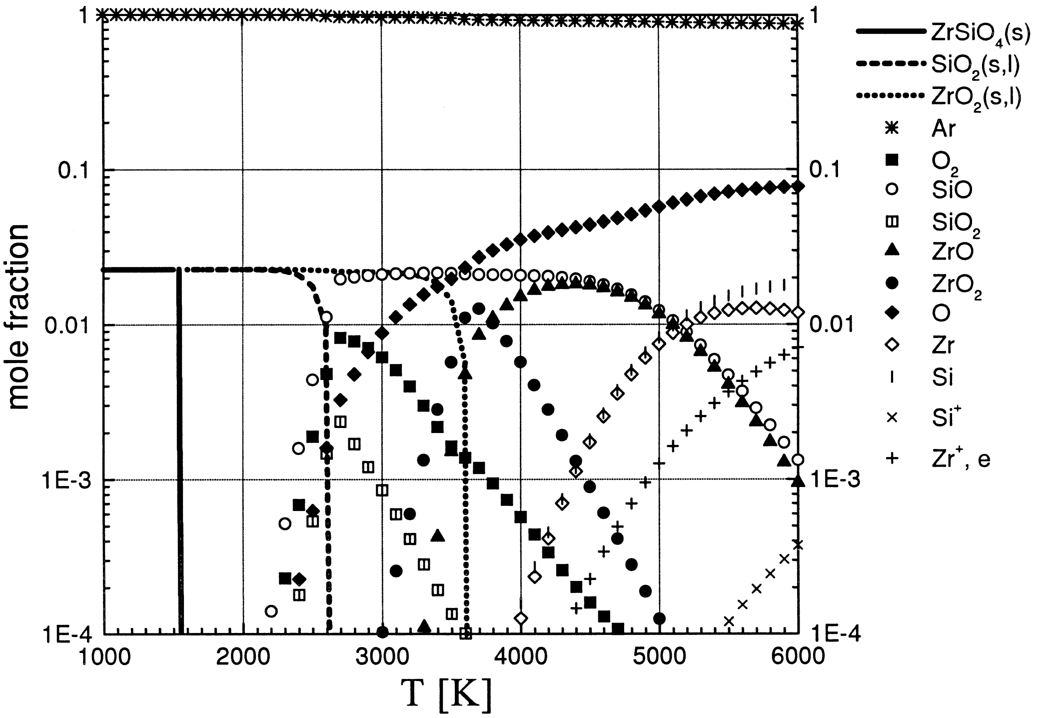

Fig. 1. Equilibrium composition of the Zr–Si–O–Ar system for Zr:Si:O:Ar = 0.02:0.02:0.08:0.88.

2. Computation method

The thermodynamic method used here, based on the Mass Action Law, is given completely in [8], and a shorter version in [9]. All nomenclature and definitions from papers [8,9] are used in this paper. The polynomial coefficients for temperature dependence of the modified free enthalpies (Gibbs energies), as enthalpies change for molecule to atoms decomposition reaction, and for heat of formation (from elements at standard state and temperature 0 K) of chemical species, are taken from data in Ref. [10], which are equivalent to those in JANAF [11], but more complete and more convenient to use. Temperature dependence of reaction constants and enthalpy for ZrSiO4(s), ZrC (s) and for ZrH (g) are compiled, and those heats of formations are taken from data in JANAF [11]. Reaction constant for ZrC (g) is taken from [2]. The ZrC(g) enthalpy temperature dependence is presumed to be same for the two-atomic gas ZrO(g) [9], and the ZrC(g) heat of formation is estimated from data for heat of formation of SiC(g) [10], SiO(g) [10] and ZrO(g) [10] (the estimated value is $\Delta H_{f,ZrC(g)} = 914.783$ kJ/mol).

3. Results and discussion

The temperature dependence of the equilibrium composition for the Zr–Si–O–Ar system with atomic ratio Zr:Si:O:Ar = 0.02:0.02:0.08:0.88 is given in Fig. 1. This system can be obtained from 0.02 mol of zircon and 0.88 mol of argon. Only three condensed phases, zircon ZrSiO4(s) for temperature $T < 1500$ K, zirconia ZrO_2 (s,l) for temperature range $1500\ K < T < 3600$ K, and silica SiO_2(s,l) for temperature range $1500\ K < T < 2550$ K, can exist, in this system. All other species are in the gaseous state. Only a small mol fraction of Zr^+ ions, Si^+ ions and electrons exists at a higher temperature (less than 1% of Zr^+ and electrons for temperature $T = 6000$ K). All presented results in the paper are related to pressure of 1 bar. Zircon dissociation to ziconia and silica in the temperature range $1400\ K < T < 1600$ K, has also been experimentally established [12].

The temperature dependence of total enthalpy per mol*[1] (including the heat of phase transformation, and the heat of formation of all chemical species in the system) for Zr–Si–O–Ar system is presented in Fig. 2. For example, 1 mol* of the system in Fig. 1 consists of 0.02 mol of Zr atoms plus 0.02 mol of Si atoms plus 0.08 mol of O atoms plus 0.88 mol of Ar atoms.

3.1. *The possibility of obtaining pure zirconium carbide powder*

First, we will consider the system with the ratio of $ZrSiO_4$ mol per number of CH_4 mol 1:4 which corresponds to chemical reaction

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[1] One mol* is defined as the quantity of the system, which consists of one mol of atoms (of all elements in the system) in total.

$$ZrSiO_4(s) + 4\ CH_4(g) \rightarrow ZrC(s) + SiO(g) + 3\ CO + 8H_2 \quad (1)$$

In Fig. 3 the temperature dependence of the equilibrium composition for the Zr–Si–O–C–H–Ar system with atomic ratio Zr:Si:O:C:H:Ar = 0.015:0.015:0.06:0.06:0.24:0.61 is shown; it can be obtained by injection of adequate quantity of methane in the system from Fig. 1. Five condensed phases, zircon $ZrSiO_4(s)$ for temperature $T < 1600$ K, graphite powder C(s) for temperature $T < 1600$ K, zirconia $ZrO_2(s)$ and silicon carbide SiC(s) for temperature range 1400 K < T < 1900 K, and zirconium carbide ZrC(s,l) for temperature range 1700 K < T < 3700 K, can exist in this system. The pure zirconium carbide powder, ZrC(s,l) can be deposited in the wide temperature interval 1950 K < T < 3700 K (Fig. 2). All other species are in the gaseous state. A small mol fraction of Zr^+ ions Si^+ ions and electrons exists at higher temperature only (less then 1% for temperature $T = 6000$ K).

The presented results indicate technological steps in a plasma chemical process for zirconium carbide powder synthesis from zircon concentrates as precursor. In the first step, zircon dissociation to zirconia and to silica in argon thermal plasma is followed by their full evaporation (Fig. 1).

In the second step by introducing methane (CH_4) and reactive quenching the temperature can be lowered to a range in which it is possible to obtain ZrC(s,l) (Fig. 3). By adequate high-temperature filtration the pure zirconium carbide powder can be separated from the system.

Example 1. The system in Fig. 1 can be obtained by injection of 0.02 mol of zircon into 0.88 mol of argon plasma. The inlet enthalpy of crystalline zircon ($ZrSiO_4$) at temperature $T = 298.15$ K is $H_{ZrSiO4}(298.15) = -1999.26$ kJ/mol [11]. Argon plasma enthalpy temperature dependence is given in Table 1. In argon plasma, it is (usually) possible to achieve (depending on plazmatron construction, heat losses to surroundings and other) the temperatures from 10,000 to 11,000 K. Enthalpy of argon plasma at temperature $T_1 = 10000$ K is $H_{Ar}(10000) = H_{s,1} = 243.5$ kJ/mol* (Table 1). With the assumption that there are no heat losses to surroundings, the enthalpy of system in Fig. 1 will be

$$\begin{aligned} H_{s,2} &= 0.02 \times H_{ZrSiO4}(298.15) + 0.88 \times H_{s,1} \\ &= 0.02 \times (-1999.26) + 0.88 \times 243.5 \\ &= 174.29\ \text{kJ/mol}^*. \end{aligned}$$

For this enthalpy value in Fig. 2 (curve 4), one can find that the corresponding temperature of the system is $T_2 > 6000$ K ($T \sim 6050$).

If we inject 0.0571 mol of cold methane, whose enthalpy at $T = 298.15$ K is $H_{CH4}(298.5) = -56.614$ kJ/

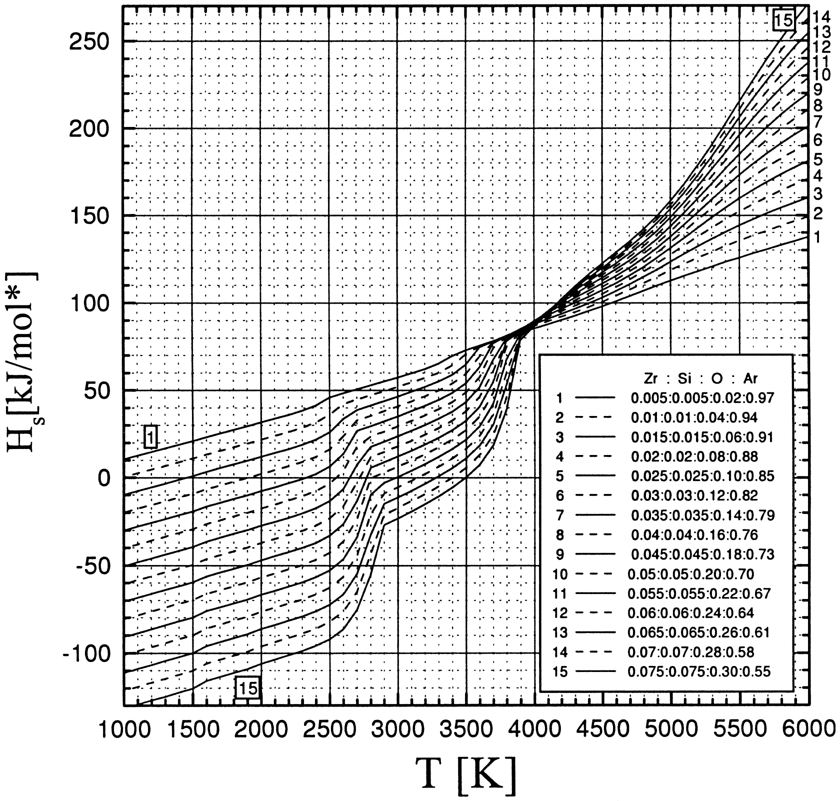

Fig. 2. Temperature dependence of enthalpy for the Zr–Si–O–Ar systems, at $p = 1$ bar.

Table 1
Enthalpy of argon plasma

T (K)	H (kJ/mol*)	T (K)	H (kJ/mol*)	T (K)	H (kJ/mol*)
1300	27.02	5900	122.7	10500	277.2
1400	29.1	6000	124.8	10600	285.2
1500	31.18	6100	126.8	10700	293.6
1600	33.26	6200	128.9	10800	302.6
1700	35.34	6300	131	10900	312
1800	37.41	6400	133.1	11000	322
1900	39.49	6500	135.3	11100	332.6
2000	41.57	6600	137.4	11200	343.9
2100	43.65	6700	139.5	11300	355.7
2200	45.73	6800	141.6	11400	368.3
2300	47.81	6900	143.8	11500	381.5
2400	49.89	7000	145.9	11600	395.5
2500	51.97	7100	148.1	11700	410.2
2600	54.04	7200	150.3	11800	425.8
2700	56.12	7300	152.5	11900	442.1
2800	58.2	7400	154.8	12000	459.4
2900	60.28	7500	157	12100	477.5
3000	62.36	7600	159.3	12200	496.5
3100	64.44	7700	161.7	12300	516.5
3200	66.52	7800	164	12400	537.4
3300	68.59	7900	166.5	12500	559.4
3400	70.67	8000	168.9	12600	582.3
3500	72.75	8100	171.5	12700	606.3
3600	74.83	8200	174.1	12800	631.3
3700	76.91	8300	176.7	12900	657.3
3800	78.99	8400	179.5	13000	684.4
3900	81.07	8500	182.3	13100	712.6
4000	83.14	8600	185.3	13200	741.8
4100	85.22	8700	188.3	13300	772
4200	87.3	8800	191.5	13400	803.2
4300	89.38	8900	194.8	13500	835.4
4400	91.46	9000	198.2	13600	868.5
4500	93.54	9100	201.8	13700	902.5
4600	95.62	9200	205.6	13800	937.4
4700	97.7	9300	209.5	13900	973
4800	99.77	9400	213.6	14000	1009
4900	101.9	9500	218	14100	1046
5000	103.9	9600	222.5	14200	1084
5100	106	9700	227.4	14300	1122
5200	108.1	9800	232.4	14400	1160
5300	110.2	9900	237.8	14500	1199
5400	112.3	10000	243.5	14600	1238
5500	114.3	10100	249.5	14700	1276
5600	116.4	10200	255.8	14800	1315
5700	118.5	10300	262.5	14900	1354
5800	120.6	10400	269.7	15000	1392

mol [10], into 0.715 of mol* of the system Zr:Si:O:Ar = 0.02:0.02:0.08:0.88 (Fig. 1) for which has been calculated enthalpy $H_{s,2} = 174.29$ kJ/mol*, the atomic content of (newly) obtained system is Zr:Si:O:-C:H:Ar = 0.0143: :0.0143:0.0571:0.0571:0.2286:0.6286.

The enthalpy of this system will be

$$H_{s,3} = 0.715 \times H_{s,2} + 0.0571 \times H_{CH4}(298.15)$$

$$= 0.715 \times 174.29 + 0.0571 \times (-56.614)$$

$$= 121.38 \text{ kJ/mol}^*.$$

For Zr:Si:O:C:H:Ar = 0.0143:0.0143:0.0571:0.0571: 0.2286:0.6286 and for enthalpy value of 121.38 kJ/mol*, in Fig. 4 (near curve 6) we can find that corresponding temperature of the system is $T_3 \approx 3750$ K. This temperature is at the upper limit of the temperature range in which it is possible to deposit pure ZrC(s) powder (Fig. 3).

The temperature dependence similar to that in Fig. 3, can be obtained using the propane–butane gas (40% propane, 60% butane mixture, with conditional molecular formula $C_{3.6}H_{9.2}$) [9] in accordance to chemical reaction

$$ZrSiO_4(s) + (10/9)C_{3.6}H_{9.2}(g) \rightarrow ZrC(s) + SiO(g) + 3\ CO + (46/9)H_2 \quad (2)$$

In Fig. 5, the temperature dependence of the equilibrium composition for the Zr–Si–O–C–H–Ar system with atomic ratio Zr:Si:O:C:H:Ar = 0.015:0.015:0.06: 0.06:0.1533:0.6967 is shown; it can be obtained by injection of adequate quantity of propane–butane to corresponding quantity of the system Zr–Si–O–Ar. Five condensed phases, zircon ZrSiO4(s) for temperature $T < 1600$ K, graphite powder C(s) for temperature $T < 1600$ K, zirconia $ZrO_2(s)$ and silicon carbide SiC(s) for temperature range 1400 K < T < 1900 K, and zirconium carbide ZrC(s,l) for temperature range 1700 K < T < 3700 K, can exist in this system. The pure zirconium carbide powder, ZrC(s,l) can be deposited in the wide temperature interval 1950 K < T < 3700 K (Fig. 2). All other species are in the gaseous state. Only a small mol fraction of Zr^+ ions, Si^+ ions and electrons exists at a higher temperature (less then 1% for temperature $T = 6000$ K).

Example 2. An iterative calculation procedure similar to that in example 1 can be conducted for reactive quenching with propane–butane gas. For this case the value of propane–butane enthalpy at $T = 298,15$ K is $H_{C3.6H9.2}(298.5) = -100.95$ kJ/mol [9], and the temperature dependence of total enthalpy for the Zr–Si–O–C–H–Ar system is given in Fig. 6.

If in the first step of the process we inject 0.0225 mol of zircon to 0.865 mol* of argon plasma. With the assumption that there is no heat losses to the surroundings, the enthalpy of the obtained system (Zr:Si:O: Ar = 0.0225:0.0225:0.09:0.865) will be

$$H_{s,2} = 0.0255 \times H_{ZrSiO4}(298.15) + 0.865 \times H_{s,1}$$

$$= 0.0225 \times (-1999.26) + 0.865 \times 243.5$$

$$= 165.64 \text{ kJ/mol}^*.$$

For this enthalpy value in Fig. 2 (between curves 4 and 5), one can find that the corresponding temperature of the system is $T_2 \approx 5700$ K.

In the second step of the process, it is necessary to inject 0.0173 mol of propane–butane ($C_{3.6}H_{9.2}$) to

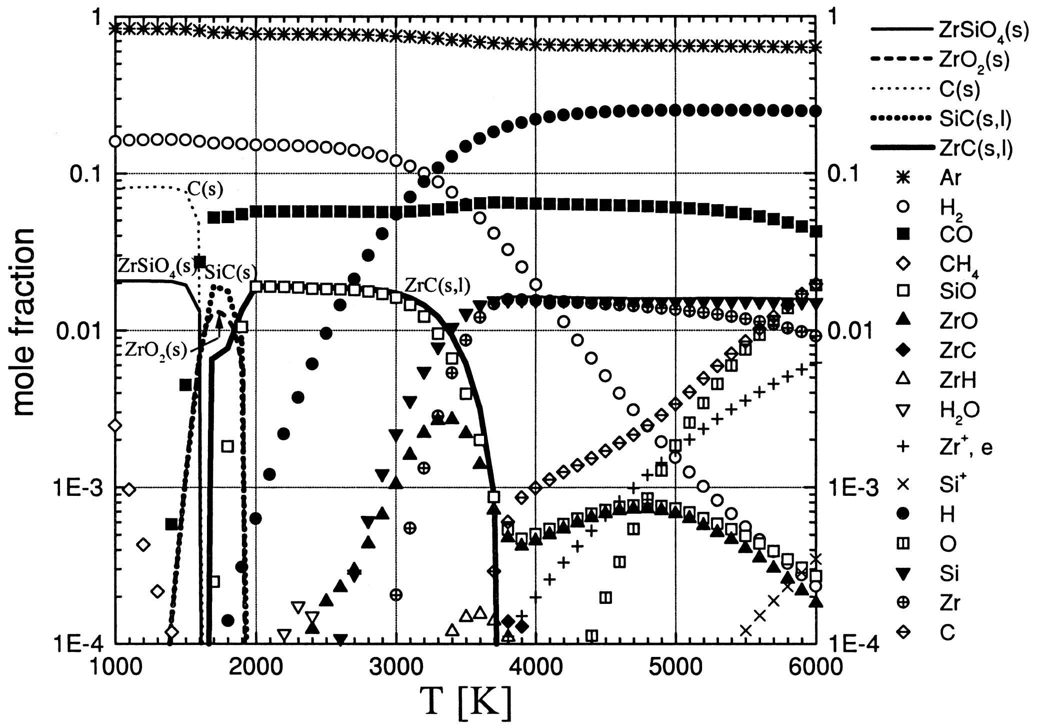

Fig. 3. Equilibrium composition of the Zr–Si–O–C–H–Ar system for Zr:Si:O:C:H:Ar = 0.015:0.015:0.06:0.06:024:0.61.

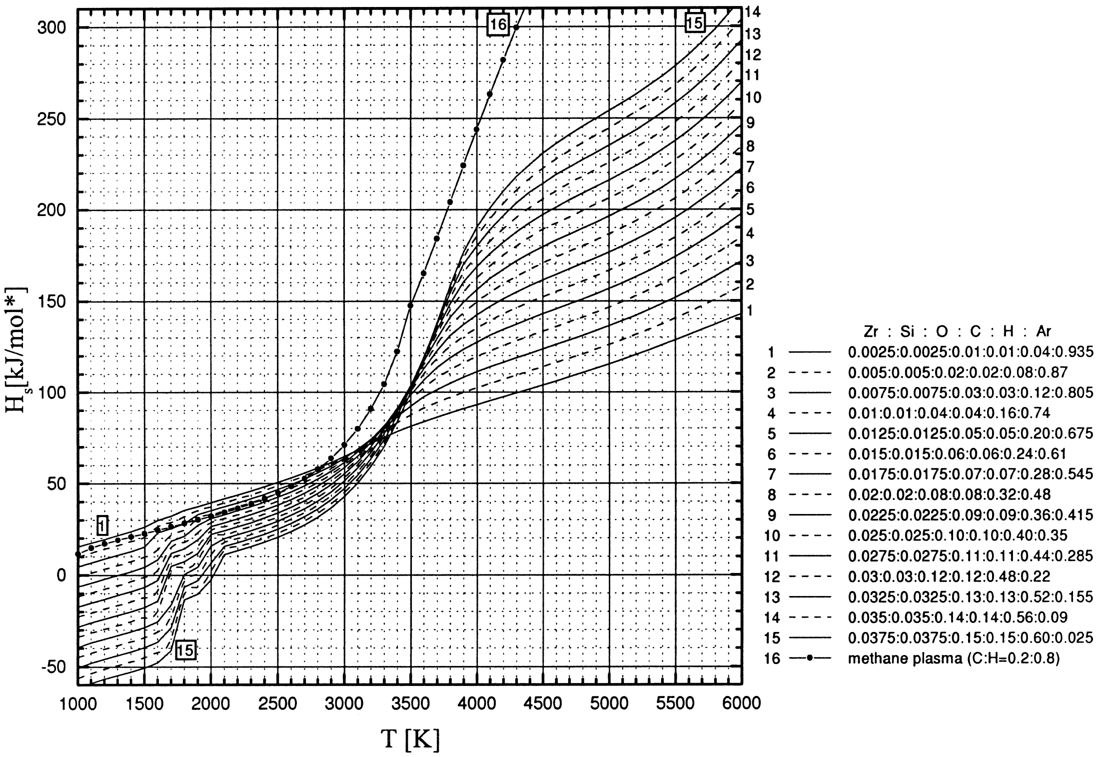

Fig. 4. Temperature dependence of enthalpy for the Zr–Si–O–C–H–Ar system, which is obtained by mixing zircon and methane (in ratio 1:4) and an adequate quantity of argon plasma.

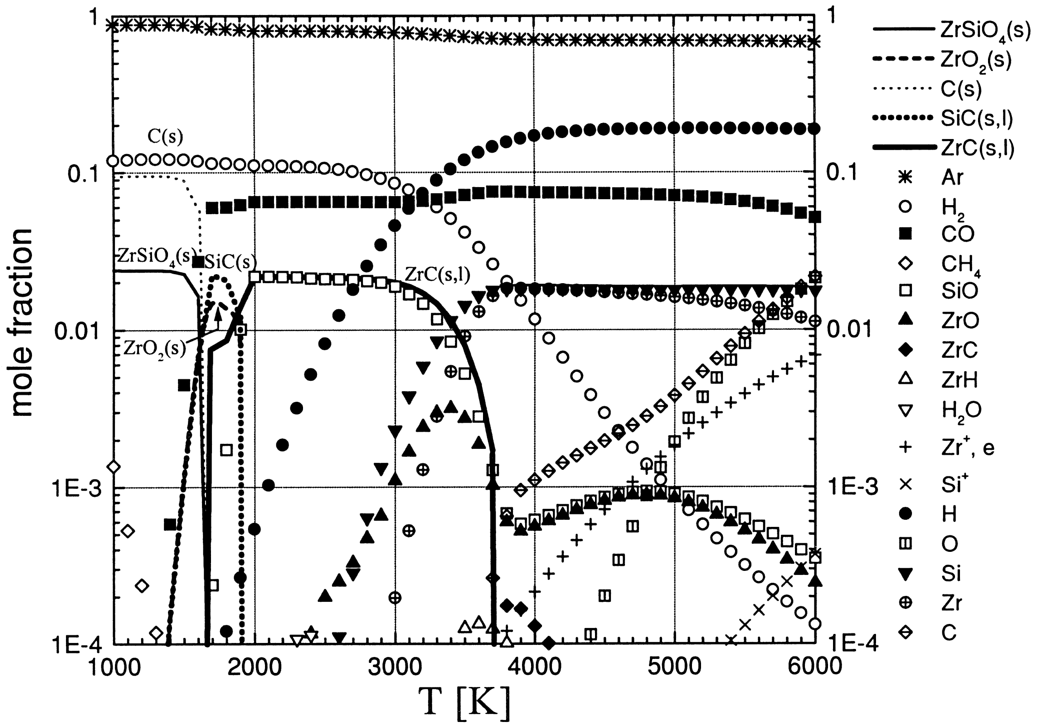

Fig. 5. Equilibrium composition of the Zr–Si–O–C–H–Ar system for Zr:Si:O:C:H:Ar = 0.0175:0.0175:0.07:0.07:01799:0.6461.

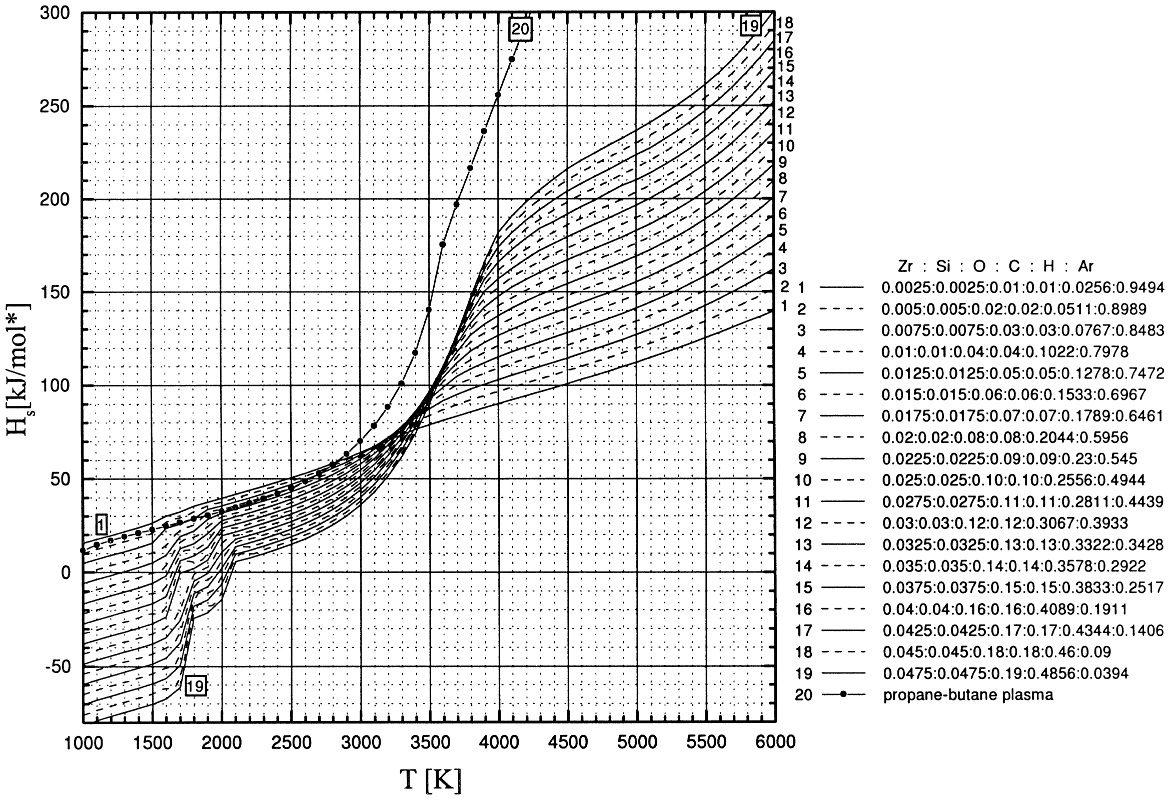

Fig. 6. Temperature dependence of enthalpy for the Zr–Si–O–C–H–Ar system, which is obtained by mixing adequate quantity of zircon, propane–butane — Eq. (2), and argon plasma.

0.7786 mol of the system in Fig. 1, to obtain 1 mol* of the system with atomic contents Zr:Si:O:C:H:Ar = 0.017:0.017:0.0682:0.0682:0.1742:0.6553.

The enthalpy of such an obtained system will be

$$
\begin{aligned}
H_{s,3} &= 0.7786 \times H_{s,2} + 0.0173 \times H_{C3.6H9.2}(298.15) \\
&= 0.7786 \times 174.29 + 0.0173 \times (-100.95) \\
&= 133.96 \ \text{kJ/mol}^*.
\end{aligned}
$$

For Zr:Si:O:C:H:Ar = 0.01557:0.01557:0.06229:0.06229: 0.1592:0.6851 and for the enthalpy value of 133.96 kJ/mol* in Fig. 6 (near curve 7) one can find that the corresponding temperature of the system is $T_3 \approx 3750$ K. This temperature is at the upper limit of the temperature range in which it is possible to deposit pure ZrC(s) powder (Fig. 5).

Procedures presented in examples 1 and 2, and data on Figs. 2, 4 and 6, can be used for processes with any other estimated, measured or determined values of heat losses to the surroundings.

Since the presented examples (1 and 2) are based on the assumption that there are no heat losses to the surroundings, they are concurrently referred to processes with a theoretical maximum of zirconium carbide powder yield per mol* of the system, which is possible to realize, if for reactive quenching, cold methane or propane–butane (at $T = 298.15$ K) are used, respectively. If we want to process greater concentrations of zircon in the system (for curves 5, 6, etc. in Fig. 2 and curves 7,8, etc. in Fig. 4 and curves 7,8, etc. in Fig. 6), it is necessary to inject the hot methane as a reactive gas in the second step of the process (through the supplemental plazmatron for example). In this case the calculation must be performed for the inlet enthalpy of hot methane or propane–butane, and for this reason the curve for methane plasma enthalpy is also presented in the Fig. 4 (curve 16) and the curve for propane–butane enthalpy is presented in Fig. 6 (curve 20). The temperature, which can be achieved with methane plasma, is 4000–4100 K and with propane–butane plasma is 4100–4200 K.

3.2. The possibility of formation of composite zirconium carbide + silicon carbide powder

In Fig. 7, the temperature dependence of the equilibrium composition for the Zr–Si–O–C–H–Ar system with atomic ratio Zr:Si:O:C:H:Ar = 0.01:0.01:0.04:0.06: 0.24:0.64 (for ratio of number of ZrSiO4 moles per number of CH_4 mol 1:6) is shown; it corresponds to the chemical reaction

$$
\begin{aligned}
ZrSiO_4(s) + 6\ CH_4(g) \rightarrow\ & ZrC(s) + SiC(s) \\
& + 4\ CO + 12H_2
\end{aligned}
\tag{3}
$$

Five condensed phases, zircon ZrSiO4(s) for temperature $T < 1600$ K, graphite powder C(s) for temperature

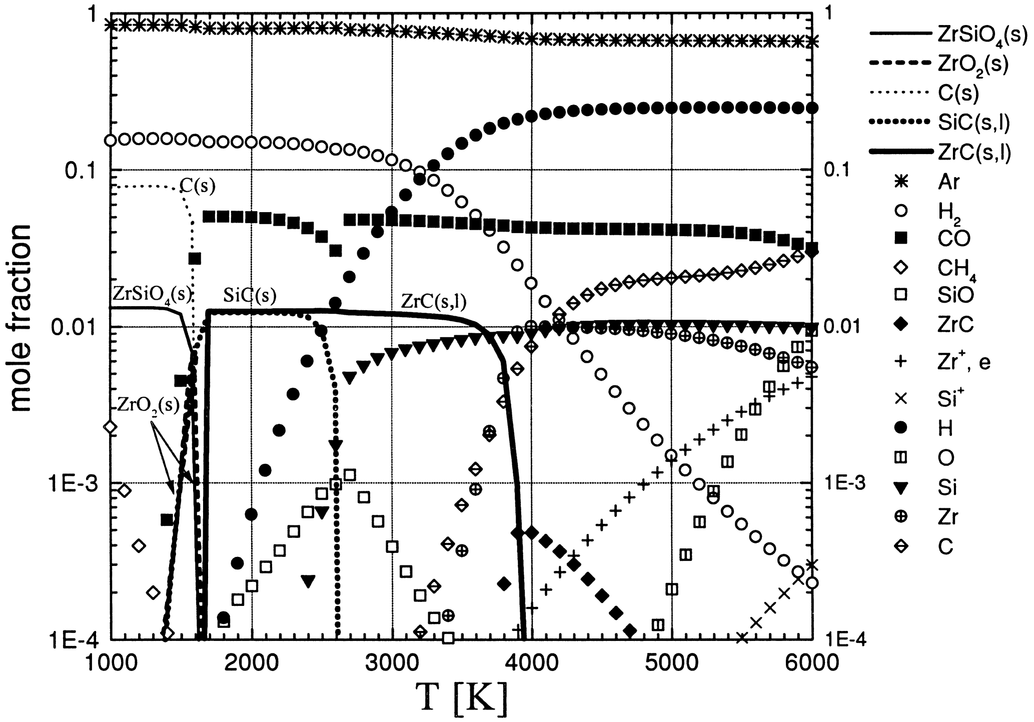

Fig. 7. Equilibrium composition of the Zr–Si–O–C–H–Ar system for Zr:Si:O:C:H:Ar = 0.01:0.01:0.04:0.06:024:0.64.

$T < 1600$ K, zirconia $ZrO_2(s)$ for temperature range 1400 K $< T <$ 1650 K, silicon carbide SiC(s) for temperature range 1400 K $< T <$ 2600 K, and zirconium carbide ZrC(s,l) for temperature range 1700 K $< T <$ 3900 K, can exist in this system. The pure zirconium carbide powder, ZrC(s,l) can be deposited in the temperature interval 2650 K $< T <$ 3900 K (Fig. 7) and composite zirconium carbide + silicon carbide powder can be deposited in the temperature interval 1700 K $< T <$ 2600 K (Fig. 7). All other species are in the gaseous state. A small mol fraction of Zr^+ ions Si^+ ions and electrons exists at higher temperature only (less then 1% for temperature $T =$ 6000 K).

In Fig. 8, the temperature dependence of the equilibrium composition for the Zr–Si–O–C–H–Ar system with atomic ratio Zr:Si:O:C:H:Ar = 0.01:0.01:0.04:0.06:0.24:0.64 is shown; it can be obtained using the propane–butane for reactive quenching in accordance with the chemical reaction

$$ZrSiO_4(s) + (10/6)C_{3.6}H_{9.2}(g) \rightarrow ZrC(s) + SiC(s) + 4\ CO + (23/3)H_2 \quad (4)$$

As we can see from Figs. 7 and 8, there is a very small difference between these two diagrams, the main difference is the smaller content of hydrogen (and corresponding substances with hydrogen) in Fig. 8.

A calculation procedure similar to that presented in examples 1 and 2 can be conducted for the systems presented in Figs.7 and 8 using the data from Figs. 2, 9 and 10. Here in example 3 we will give the calculation of a one step process (cocurrent injection of zircon and quenching gas into argon plasma) with presumption that heat losses to the surroundings Q are 40% of total argon plasma enthalpy [$Q = 0.4H_{Ar}(10000) = 0.4H_{s,1} = 0.4243.5 = 97.4$ kJ/mol*].

Example 3. To obtain the system from Fig. 7, it is necessary to inject 0.01 mol of zircon and 0.06 mol of methane (CH_4) into 0.64 mol* of argon plasma. If heat losses to the surroundings are $Q = 97.4$ kJ/mol*, the enthalpy of such an obtained system will be

$$\begin{aligned} H_{s,3} &= 0.01 \times H_{ZrSiO4}(298.15) + 0.06 \times H_{CH4}(298.15) \\ &\quad + 0.64 \times H_{Ar}(10\,000) - Q \\ &= 0.01 \times (-1999.26) + 0.06 \times (-56.614) + 0.64 \\ &\quad \times 243.5 - 97.4 = 35.05\ \text{kJ/mol}^*. \end{aligned}$$

For this enthalpy value (35.05 kJ/mol*) and for Zr:Si:O:C:H:Ar = 0.01:0.01:0.04:0.06:0.24:0.64 in Fig. 9 (curve 4) we can find the temperature of the system $T_3 \approx$ 2200 K. At this temperature (Fig. 7) it is possible to obtain composite zirconium carbide + silicon carbide powder.

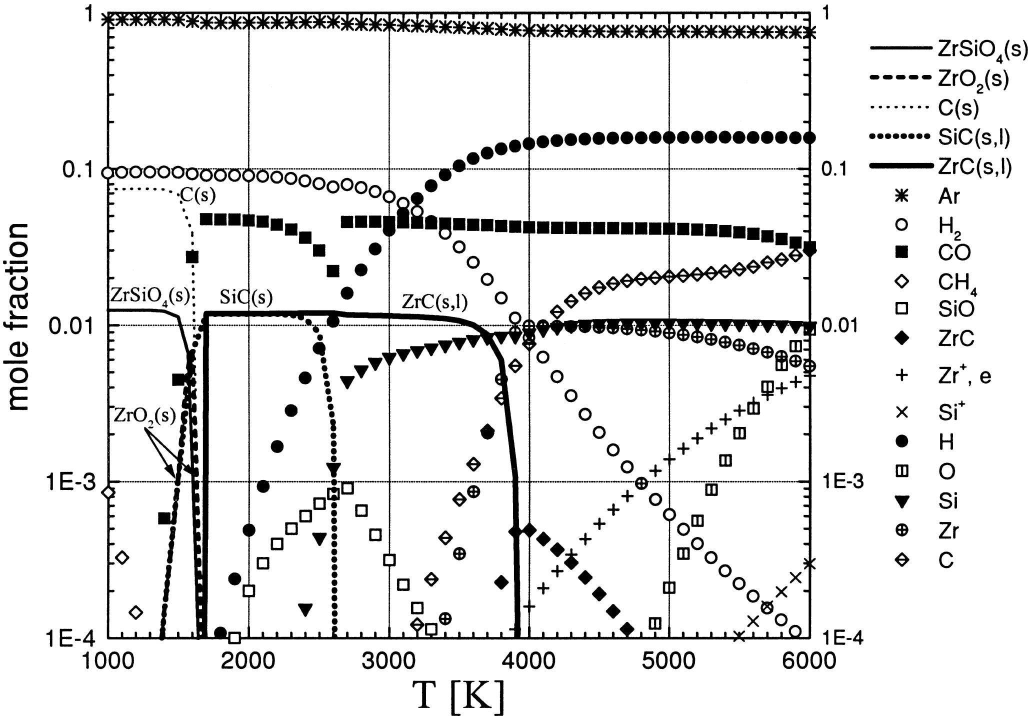

Fig. 8. Equilibrium composition of the Zr–Si–O–C–H–Ar system for Zr:Si:O:C:H:Ar = 0.01:0.01:0.04:0.06:01533:0.7267.

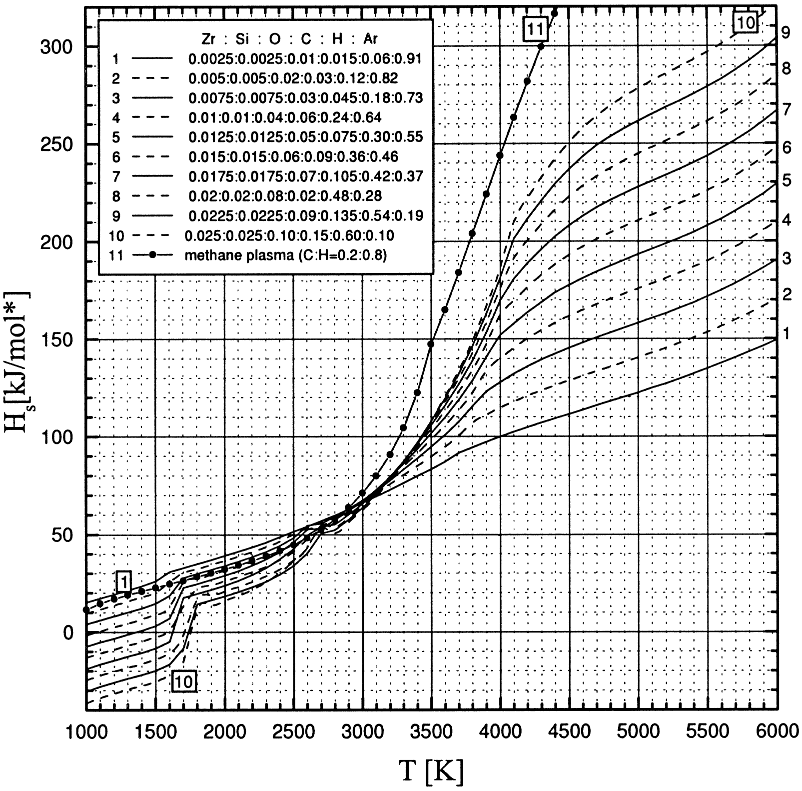

Fig. 9. Temperature dependence of enthalpy for the Zr–Si–O–C–H–Ar system, which is obtained by mixing zircon and methane (in ratio 1:6) and adequate quantity of argon plasma.

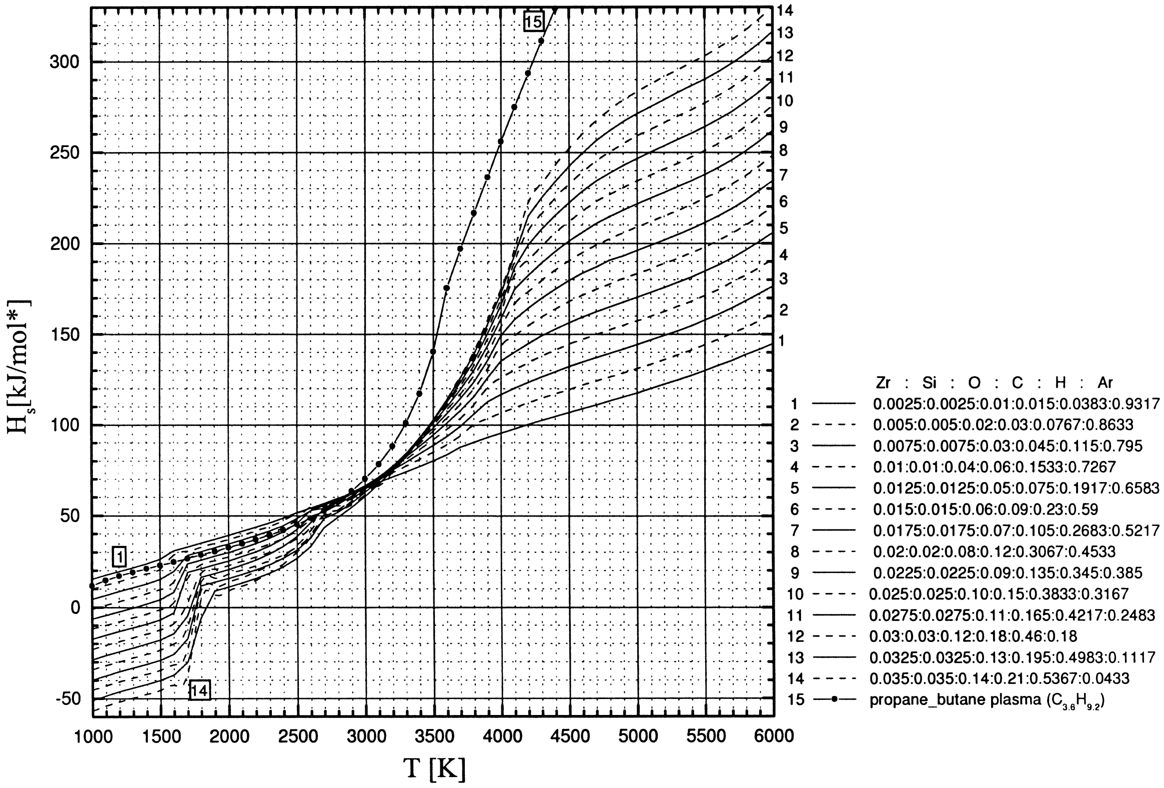

Fig. 10. Temperature dependence of enthalpy for the Zr–Si–O–C–H–Ar system, which is obtained by mixing adequate quantity of zircon, propane–butane — Eq. (4), and argon plasma.

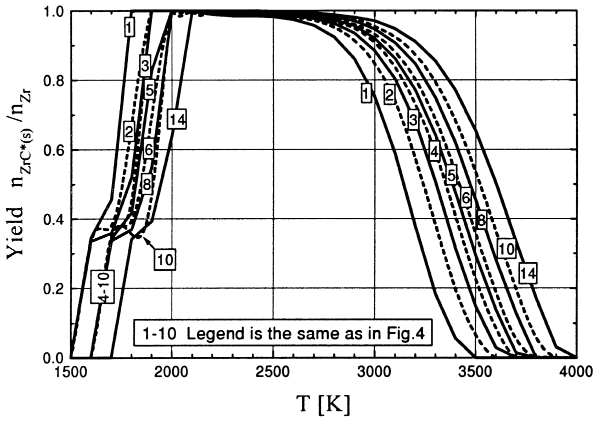

Fig. 11. The yield in the systems according to Eq. (1).

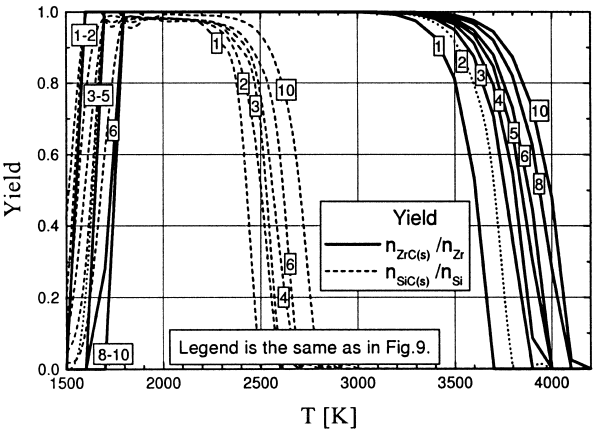

Fig. 13. The yield in the systems according to Eq. (3).

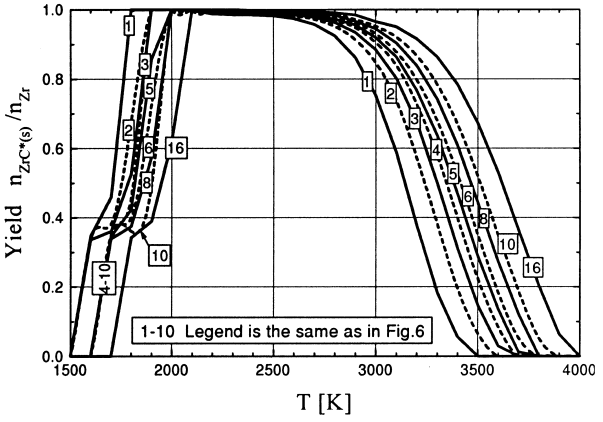

Fig. 12. The yield in the systems according to Eq. (2).

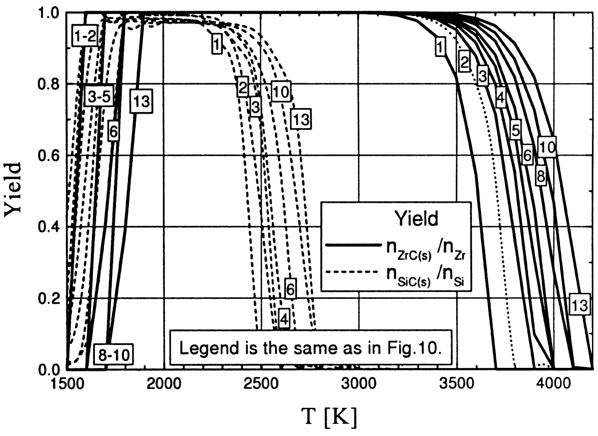

Fig.14. The yield in the systems according to Eq. (4).

To obtain the system from Fig. 8, it is necessary to inject 0.01 mol of zircon and 0.06/3.6 mol of propane–butane ($C_{3.6}H_{9.2}$) into 0.7267 mol* of argon plasma. The same procedure for the system in Fig. 8, [for $H_{C3.6H9.2}(298.15) = -100.95$ kJ/mol and for $Q = 0.3243.5 = 73.05$ kJ/mol*] gives

$$\begin{aligned} H_{s,3} &= 0.01 \times H_{ZrSiO4}(298.15) + 0.06/3.6 \\ &\quad \times H_{CH4}(298.15) + 0.64 \times H_{Ar}(10\,000) - Q \\ &= 0.01 \times (-1999.26) + 0.06/3.6 \times (-100.95) \\ &\quad + 0.7267 \times 243.5 - 73.05 = 82.23 \ \text{kJ/mol}^* \end{aligned}$$

and $T_3 \approx 3300$ K (Fig. 10 curve 4). In Fig. 8 it can be seen, that in this case, pure zirconium carbide powder can be deposited from the system.

3.3. Yield

The yield [number of ZrC(s) mol per number of Zr mol in the system [or number of SiC(s) mol per Si mol, respectively] for considered cases [Eqs. (1)–(4)] is presented in Figs. 11–14.

4. Conclusion

The presented results of thermodynamic computation (of the equilibrium composition and total enthalpy behavior) of the Zr–Si–O–Ar and Zr–Si–O–C–H–Ar systems enables the determination and optimization of the temperature, mass and enthalpy parameters for the processing of zirconium carbide powder production from zircon in argon thermal plasma. The method of process parameter determination and optimization is illustrated by two examples: for reactive quenching with cold methane or with cold propane–butane gas, in the second step of the process.

In the first step of the process, zircon dissociation to zirconia and silica in argon thermal plasma is followed by their full evaporation. As a result of this process, the argon plasma partially cools, and (with products of zircon) gives the Zr–Si–O–Ar system. In the second step of the process, sufficient quantity of methane or propane-butane gas (as reduction and cooling reagents) is injected in the system of Zr–Si–O–Ar; this process gives adequate Zr–Si–O–C–H–Ar system and, in the corresponding temperature range, zirconium carbide powder.

The systems from which it is possible to obtain composite zirconium carbide + silicon carbide powder, are also considered in the paper.

Presented results can also be used for one step process[2] parameters calculation and optimization.

Generally, the results of such calculations enable cutting down experimental work and decreasing the cost of investigations.

References

[1] L.E. Toth, Refractory Materials, Vol. 7. Transition Metal Carbides, Academic Press, New York, NY, 1971, pp. 4, 6, 9–11).

[2] I.S. Kulikov, Termodinamika karbidov i nitridov, (spravochn), Metalurgiya, Chelyabinsk, 1988, p. 110.

[3] G.V. Samsonov, Plenum Press Handbook of High Temperature Materials, No. 2: Properties Index, Plenum Press, New York, NY, 1964, pp. 336–337.

[4] W.B. Johnson, et al., Kinetics of formation of a plated-reinforced ceramic composite prepared by the direct reaction of zirconium and boron carbide, J. Am. Ceram. Soc. 74 (9) (1991) 2093–2101.

[5] E. Breval, W.B. Johnson, Microstructure of plated-reinforced ceramic composite prepared by the direct reaction of zirconium and boron carbide, J. Am. Ceram. Soc. 75 (8) (1992) 2139–2145.

[6] E.E. Vidal, P.R. Taylor, M. Manrique, Synthesis of zirconium carbide from zircon concentrates, in: R.Y. Lin, Y.A. Chang, R.C. Liu, C.T. Liu (Eds.), Design Fundamentals of High Temperature Composites, Intermetallics and Metal–Ceramics Systems, The Minerals, Metals & Materials Society, USA, 1995, pp. 345–356.

[7] Banqui Wu, P.R. Taylor, E.E. Vidal, in: B. Mishra, (Eds.), Zirconium Carbide Synthesis in a High Temperature Plasma Reactor, EPD Congress 1997, The Minerals, Metals & Materials Society, 1997, pp. 245–254.

[8] Ž.G. Kostić, P.Lj. Stefanović, P.B. Pavlović, Ceram. Int. 22 (3) (1996) 179–186.

[9] Ž.G. Kostić, P.Lj. Stefanović, P.B. Pavlović, D.B. Cvetinović, S.Đ. Šikmanović, Ceram. Int. 26 (2) (2000) 165–170.

[10] V.P. Glushko, L.V. Gurevich (Eds.), Termodinamicheskie Svoistva Individualnyikh Veshchestv, Nauka, Moskva, (T.I) 1978, (T.II) 1979, (T. III) 1981.

[11] D.R. Stull, H. Prophet, (Pr. Dir.) JANAF Thermochemical Tables, 2nd Edition, NSRDS-NBS 37, USA, June 1971.

[12] R. McPherson, B.V. Shafer, Spherulites and phase separation in plasma-dissociated zircon, J. Mater Sci. 19 (1984) 2696–2704.

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[2] Simultaneous injection of zircon concentrates and quenching reagents (methane or propane–butane gas) in argon thermal plasma.