Functional Materials 11, No.4 (2004)

Plane-elliptical mirror furnace for crystal growth

S.Novaconi, Al.Boltosi, R.Baies, M.Bartan*, I.Grozescu

National R&D Institute for Electrochemistry and Condensed Matter,
Timisoara, Romania
*Exotic Materials Research and Tehnology, Inc., Cleveland, Ohio, USA

Received November 25, 2003

For single crystal growing in aggressive media, classical heating methods do not provide reliable results. In this study, we conceived and realized a system for radiant heating, by which the growth media is separated by the radiant and radiation focusing elements. The system is composed from an assembly of plane-elliptical mirrors with rod-shaped halogen lamps in the first focal centers and crucible as the second focal center. The growth medium being isolated, we could employ highly oxidant atmospheres and use crucibles made of dielectric oxide materials. The results obtained show a possibility to use the system to synthesize materials in aggressive media, as well as to configure the specific temperature gradients required by these processes.

Классические методы нагрева не обеспечивают надежных результатов при выращивании монокристаллов в агрессивных средах. В этом исследовании нами разработана и реализована система для радиационного нагрева, с помощью которой достигается отделение ростовой среды от излучающих и фокусирующих излучение элементов. Система состоит из плоско-эллиптических зеркал с галогеновыми лампами стержневидной формы в первых фокальных центрах и тиглем во втором фокальном центре. Поскольку ростовая среда изолирована, возможно применение сильно окислительных сред и использование тиглей из диэлектрических оксидных материалов. Полученные результаты иллюстрируют возможность применения системы для синтеза материалов в агрессивных средах, а также для конфигурирования специальных систем градиентов температуры, необходимых для таких процессов.

The preparation of materials in aggressive and controlled media at elevated temperatures is usually difficult to realize, especially due to the requirements to the precursor material heating system. The classical methods are based on the employing of heating resistances and/or heating by induced currents. However, it is known that above a certain temperature and in the presence of oxygen atmosphere, the resistors are dramatically affected by different reactions that take place in this environment. Besides, the heating by induced currents requires the use of crucible made of conductive materials, but almost all metals prone to react at elevated temperatures and in the presence of oxygen. When working at temperatures above 1600°C as we are, the only option is to use platinum, iridium or iridium-platinum alloy crucibles which, however, increases the process cost. Therefore, one of the most advantageous method is the indirect heating by focusing of the heat radiation. Heating systems based on the radiation focusing of the light arising from halogen lamps using elliptical mirrors have been developed and used for materials processing [1, 2]. Recently, studies upon the use of Xenon discharge lamps have been reported [3] and heating processes as well as temperature distribution were evaluated for different furnace types [4–6]. Nowadays, efforts are given to the development and implementation of installation for material

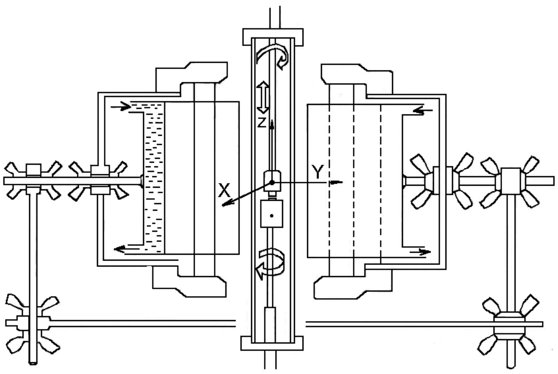

Fig. 1. Radiant heating system.

growing with separation of media [7, 8] as well as for various investigations in space [9].

We present here the installation for crystals growing from melt in a controlled medium, assisted by the presence of oxygen as the growing atmosphere. By employing Czochralski method for material growing in dielectric oxide-based crucible, an uniform temperature distribution and additionally the appropriate gradient required by the growth processes should be realized. Our approach aims to implement an heating system based on focusing of the radiation emerging from halogen lamps using plane-elliptical mirrors. Although this method exhibits a lower efficiency than that obtained with elliptic mirrors, it enables to achieve a high longitudinal temperature uniformity over the external crucible surface. The temperature gradients necessary in the growing processes were obtained by the use of deflectors at defined positions in the system.

The chamber for growth process is made of a quartz tube into which the crucible containing the precursor material, the rotating support and the seed with its corresponding device are placed (Fig. 1). The rotation system of the crucible together with the devices providing rotating and axial movement of the seed are placed outside the chamber. All movements and alignments can be controlled and independently adjusted. The movement transmission systems are tightened. The growth chamber is connected to a vacuum pump and then to a system for quantifying and mixing the gases. The heating system with radiant and focusing elements is placed outside the growth chamber. The heating system is composed of heating elements each consisting of a plan-elliptical mirror and halogen lamp of rod shape placed in the first focal center.

The mirrors are made from 1 mm thick brass plate coated with a reflecting layer, followed by coating with a protective thin chrome layer. For the reflecting layer, only materials with an increased reflection in radiation spectrum of the halogen lamp are used. In this regard, the most suitable materials (Fig. 2) are aluminum, gold, and silver. Silver has been chosen in our study due to its increased reflection of about 95 to 98 % (Table 1) over the entire visible spectrum, but also because of its low cost. The focusing on the crucible has been performed in axial plane.

We studied the temperature variation as a function of time (Fig. 3) in the second focal center of the plan-elliptical mirror. Nominal voltage has been delivered for all

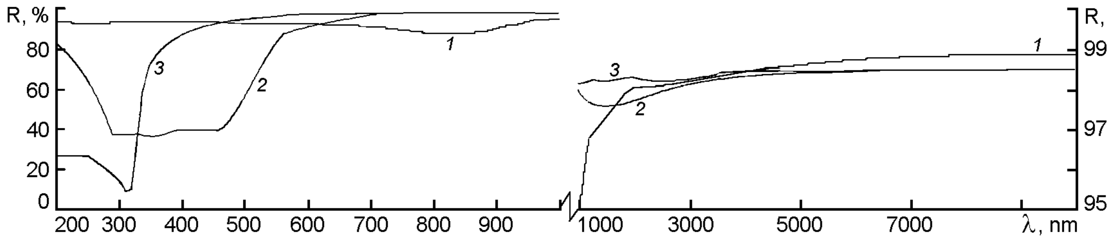

Fig. 2. Reflectance in UV/VIS/NIR for aluminum (*1*), gold (*2*) and silver (*3*).

Table 1.

Metals	Average Refl % Vis / IR	Regions of high absorption	Comments
Aluminum	92/98	700–950 nm	Sensitive deposition parameters necessary to prevent "Blueing" (scatter in visible). Thin layer of Al_2O_3 is formed on surface
Gold	94/98	300–550 nm	Adhesion problems with glass. Very soft surface. Use chrome as interlayer
Silver	95/98	UV	Tarnishing problems. Very soft surface.

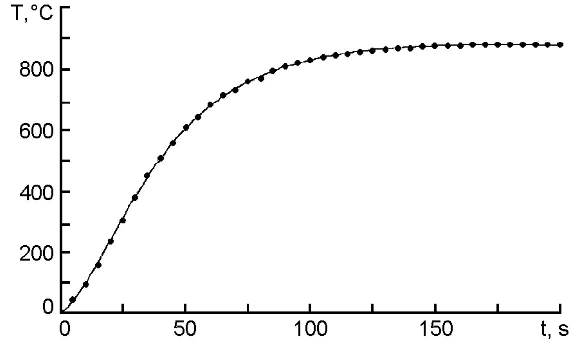

Fig. 3. Time-temperature diagram in the second focal center of the plan-elliptical mirror.

systems. The material placed in the second focal center has the absorption index of 52 % in visible spectrum, mass of 2 g and the enthalpy of 27.3 J/K·mol.

The temperature in the second focal center as a function of the applied voltage to the system formed from one and two respectively mirror-bulb assemblies is shown in Fig. 4. The plane-elliptical mirrors have the height of 80 mm, F1 at 20, F2 at 70 mm, and the angle seen by the lamp in the ellipse plane is 220°. The temperature increases approximately linearly in both cases, only the slope is different.

The temperature distribution along an axis parallel with the lamp, which intersects the second focal plan, for the configurations with 1 and 2 radiant systems is shown in Fig. 5. In comparison with distributions achieved with elliptic reflectors [10], the temperature is constant on a higher level (plateau), thus advantageous for the crystal processing in dielectric

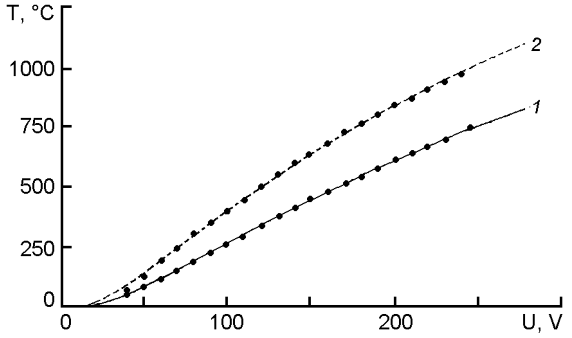

Fig. 4. Temperature-voltage diagram the system formed from one (*1*) and two (*2*) respectively mirror-bulb assemblies.

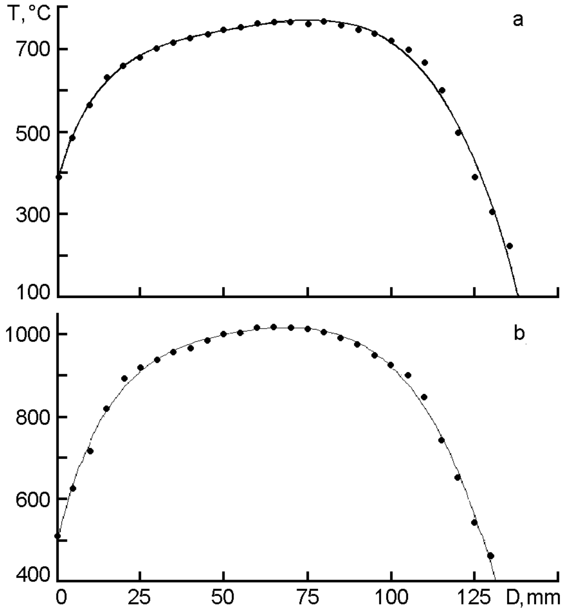

Fig. 5. Axial temperature profile at different location for 1 (a) and 2 (b) systemes.

Table 2.

Parm	Value	Std Error	t-value	99 %	Confidence Limits
a	382.4484583	11.54094602	33.13839763	350.0504151	414.8465015
b	26.19311012	1.669425425	15.68989529	21.50665574	30.87956451
c	−1.31955167	0.109364279	−12.0656551	−1.62656191	−1.01254143
d	0.189923683	0.015783010	12.03342586	0.145617207	0.234230159
e	−0.00808603	0.000638559	−12.6629352	−0.00987861	−0.00629345
Respective					
Parm	Value	Std Error	*t*-value	99 %	Confidence Limits
a	494.5650218	11.71873239	42.20294530	461.5343202	527.5957234
b	34.03791277	1.763672665	19.29945020	29.06678248	39.00904306
c	−1.55413098	0.120150618	−12.9348563	−1.89279041	−1.21547155
d	0.215307557	0.017675718	12.18097932	0.165486352	0.265128762
e	−0.00896772	0.000728888	−12.3032990	−0.01102218	−0.00691326

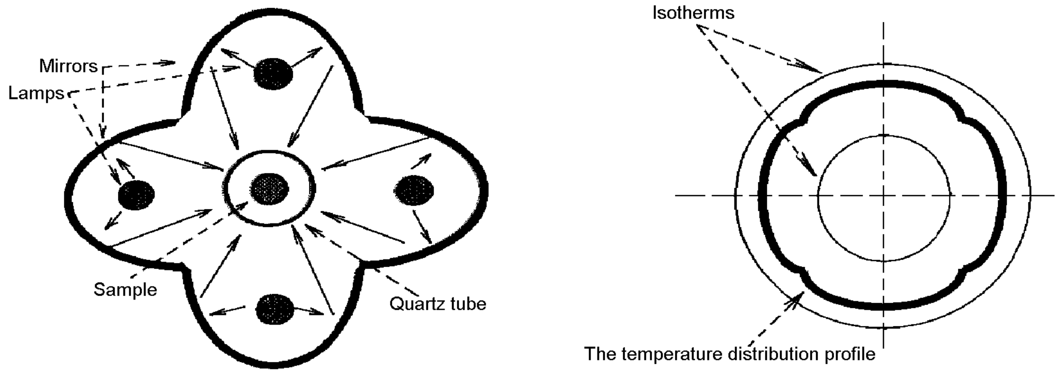

Fig. 6. Schematical design of the optical furnace with four mirrors and distribution curve of the temperature.

oxide-based crucible even if the efficiency is lower.

For the two above described cases the equation constants presented in Table 2.

The defocusing occurs due to the mirror shape deviation, implicitly of focusing determined by dilatation. This has been minimized by the use of a forced cooling system with air currents and additionally by using filament lamps of maximum available diameter. At four radiant systems, the temperature distribution in the focal center is more uniform, especially the radial one (Fig. 6). The use of a greater number of systems gives rise to unjustifiable mechanical and designing problems.

To conclude, the optical furnace with plane-elliptic mirrors is recommended for melting growth materials in aggressive atmosphere. As radiant elements, rod-shaped halogen lamps could be employed. As compared to furnaces with elliptic mirrors, the axial distribution of the temperature is much more uniform. A deviation from the plane-elliptical mirror shape affects drastically the temperature obtained in the second focal center. The lower efficiency affects somewhat the working temperatures, however, this has been overcame by increasing the power. The cooling of the assemble mirror-lamp can be effected using cold air currents. The temperature gradients are easily realizable by defined alignment and positioning of the radiation deflectors.

Acknowledgements. We thank Ministry for Education, Research and Youth for the entire support provided within the National Program MATNANTECH granted to the Institute for Research and Development for Electrochemistry and Condensed Matter, Timisoara.

References

1. H.G.Riveros, W.K.Cory, R.Toca, E.Camarillo, *J.Cryst. Growth*, **49**, 85 (1980).
2. J.G.Bednorz, H.Arend, *J.Cryst. Growth*, **67**, 660 (1984).
3. C.W.Lang, J.C.Leu, Y.H.Uang, *Cryst.Res. Technol.*, **35**, 167 (2000).
4. Rivas, C.Vazquez-Espi, *J.Cryst. Growth Adv. Space Res*, **29**, 575 (2002).
5. K.Kitazawa, K.Nagashima, T.Mizutani et al., *J.Cryst. Growth*, **39**, 211 (1977).
6. D.Rivas, C.Vazquez-Espi, *J.Cryst. Growth*, **223**, 433 (2001).
7. A.Eyer, R.Nitsche, H.Zimmerman, *J.Crystal Growth*, **47**, 219 (1979).
8. A.Eyer, B.O.Kolbsen, R.Nitsche, *J.Cryst. Growth*, **57**, 145 (1982).
9. A.Boltosi, S.Novaconi, I.Grozescu, Invention Certificate, No.A/00553 (2003).

Піч з плоско-еліптичними дзеркалами для вирощування монокристалів

С.Новаконі, Ал.Болтозі, Р.Байс, М.Бартан, І.Грозеску

Класичні способи нагрівання не забезпечують надійних результатів при вирощуванні монокристалів в агресивних середовищах. У цьому дослідженні розроблено та реалізовано систему для радіаційного нагріву, за допомогою якої досягається відділення ростового середовища від випромінювальних та фокусувальних елементів. Система складається з плоско-еліптичних дзеркал з галогеновими лампами стрижневої форми у перших фокальних центрах та з тиглем у другому фокальному центрі. Оскільки ростове середовище ізольоване, можливим є застосування сильно окиснювальних середовищ та використання тиглів з діелектричних оксидних матеріалів. Отримані результати посвідчують можливість застосування системи для синтезу матеріалів в агресивних середовищах, а також для формування спеціальних систем градієнтів температури, необхідних для таких процесів.