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ANALYTICAL SOLUTIONS FOR THERMAL TRANSIENT PROFILE IN SOLID TARGET IRRADIATED WITH LOW ENERGY AND HIGH BEAM CURRENT PROTONS

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ABSTRACT

There were obtained analytical solutions for thermal transient in solid targets, used in short half-life radioisotopes production, when irradiated with low energy and high beam current protons, in the cyclotron accelerator Cyclone 30 of the Institute for Energy and Nuclear Research (IPEN/CNEN-SP). The beam spatial profile was considered constant and the time depended heat distribution equation was resolved for a continuous particles flow entering the target. The problem was divided into two stages: a general solution was proposed which is the sum of two functions, the first one related to the thermal equilibrium situation and the second one related to a time dependent function that was determinate by the setting of the contour conditions and the initial conditions imposed by the real problem. By that one got an analytic function for a complete description of the heat transport phenomenon inside the targets. There were used both, numerical and symbolic computation methods, to obtain temperature maps and thermal gradients and the results showed an excellent agreement when compared with purely numerical models. The results were compared with obtained data from Gallium-67 and Thallium-201 irradiation routines conducted by the IPEN Cyclotrons accelerators center, showing excellent agreement. The objective of this paper is to develop solid targets irradiation systems (metals and oxides) so that one can operate with high levels of current beam, minimizing the irradiation time and maximizing the final returns.

1. INTRODUCTION

According to the International Atomic Energy Agency (IAEA)[1] in 2006, there were 262 cyclotrons accelerators operating in 39 countries in its membership. Growth of 7% compared to 246 cyclotrons accelerators in operation reported in 2002, all geared to routine production of radionuclides of medical interest.

It is believed that there are, nowadays, about 350 of these installations operating or being installed around the globe. This growth in cyclotrons in operation over the past 6 years is due to several factors: the advancement of technology for production of clearer and detailed images, the introduction of compact cyclotrons, often called "Baby's," which, besides the relative low cost, they are easy to operate and maintain, and finally, the rapid development of Positron Emission Tomography/Computing Tomography.

The number of commercially available cyclotrons is relatively large and is also growing due to the increasing demand for neutron-deficient radioisotopes of short half-life and biologically compatible.

Most of the currently installed accelerators work with energy between 10 and 30 MeV, average current of 75 µA for a period of 25 hours per week or more.

Because of the sharp growth in global demand for cyclotron radioisotopes, many centers have increased considerably the number of weekly irradiation hours. Moreover, this procedure presents some prohibitive factors the biggest one is the stopping time for corrections or preventive maintenance.

Another way to find this demand growth has been accelerating the acquisition of new and more modern ones. However it also presents obstacles, especially for the poorest and in development countries, because of the high acquisition, installation and operation costs.

Attentive to these factors, some research lines have suggested increasing the number of particles in each incident radiation, in other words, the increase of the nominal values of beam current employed, which is directly related to the final production yield of each radioisotope.

However, one should have in mind the thermal power factor transferred to the target, which is directly affected by the increase of the beam current, the temperature of the target must be kept below certain values to avoid losing material by sublimation or even the target destruction.

Commercially, what one should do in these cases is to build a super-sized refrigeration system to ensure an efficient heat exchange between the target and the coolant.

2. OBJECTIVE OF THIS WORK

In Cyclotron Accelerators Management that belongs to the Directorate of Radiopharmaceuticals (DIRF) of the Institute of Energy and Nuclear Research (IPEN), the Government of São Paulo State and managed by the National Commission for Nuclear Energy (CNEN), are installed two cyclotron accelerator types:

1. Cyclone 30, manufactured in Belgium by "Ion Beam Applications – IBA", accelerates H^- ions and extracts H^+ ions with variable energy between 15 and 30 MeV and beam current up to 0.35 mA. It has two external lines of beam and is currently employed in routine production of Iodine-123, Gallium-67 and thallium-201.

2. Cyclone-18, manufactured in Belgium by "Ion Beam Applications - IBA, accelerates H^- ions and extract H^+ ions with fixed energy of 18 MeV and maximum current of 0.15 mA (single beam) or 0.075 mA (double beam). It has 8 fixed targets disposed diametrically opposite, two by two. It is used in routine production of Fluorine-18.

The second accelerator was purchased by IPEN to fill all the current demand by ^{18}F-FDG, but may also receive at least one external line of beam to produce other radioisotopes, which have large cross section to the activation between 9 and 15 MeV , such as ^{124}I, which has been very efficient in various production techniques of the brain and thyroid PET images, or the ^{64}Cu produced by irradiation of ^{64}Ni, excellent images of the brain blood flow and also for specific myocardium diseases diagnostics.

In view of the interest of raising the income production with the increase of the nominal values of beam current, this work aims to present a mathematical-physical model to the energy transferred between the beam particles and the constituent target material, focusing in the main transient regime when the target temperature distribution is extremely uneven, displaying intense gradients.

In the first approach, this paper analyzes and presents results only for targets made of solid materials such as metals and their alloys and oxides in general.

When irradiated solid targets with beams of protons with low energy (<30 MeV) and high-current beam intensities (> 0.2 mA), there are many aspects that must be carefully analyzed and understood, such as the target geometry, its density, thermal capacity and thermal conductivity, the incident beam spatial profile , among others.

The interaction of an energetic particles beam with matter in general produces a relatively high density of thermal power (MW/m^2). Roughly speaking, the thermal power transferred to the target by an accelerated particles beam with kinetic energy E (MeV) and beam current I (μA) is given by the expression:

$$P \approx E \times I \quad \text{(Watt)} \tag{1}$$

Taking into account the target geometry, that in most of the practical cases, presents a frontal projection of approximately circular shape of radius 0.5 cm, the area exposed to the beam will be somewhere around 0.8 cm^2 and with this density thermal power directly into the target surface shall be 75 MW/m^2, depending on the intensity of the beam current.

Considering now the thickness of the target and its thermal conductivity may be inferred from the temperature gradients caused by the sudden deposition of energy on the target surface. Knowing that the power per unit area for a continuous energy flow can be written as:

$$\frac{P}{A} = \frac{K \times \Delta T}{e} \tag{2}$$

Where "P" is the power in Watt, "A" is the area in cm^2, “K” is the thermal material conductivity in W / m.K and "e" the thickness of the material. Considering that most of the solid targets used in production are about 0.5 cm thick, one can calculate the gradients of temperature for the material most commonly used. Table 1 provides some of these results, considering an average power of 2 kW.

F. M. Nortier and colleagues [2] made a study on the solid targets thermal behavior for radioisotopes production with energy protons below 30 MeV and beam current up to 0.46 mA. For the two of them there was employed two commercial solid targets capable of 7.5 kW and 15 kW, respectively, withstanding thermal power.

In their experiments, there was been modified both, the spatial profile of the beam, which directly affects the distribution of power radiated per unit area, and the flow of coolant. The results showed that the target temperature was not higher than 140 ºC for a minimum flow of 10 l / min. of water.

Table 1. Gradients of temperature for various types of Materials irradiated with a power density of 6 kW/cm^2.

Material	$K(W/m.K)$	$\Delta T(Kelvin)$
Silver	406	308
Cooper	385	324
Aluminum	205	609
Oxides	~ 0.5	25,000

W. L. Talbert and colleagues [3] conducted a study which took into account the aspects of design and construction of targets for high thermal power, R. A. Pavan and colleagues [4] have a job modeling for the thermal state of solid targets designed to withstand high beam intensities of current and J. J. Čomor and collaborators [5] built a model of the thermal properties of tellurium oxide (TeO_2) to produce ^{124}I. This paper examines aspects of heat transference to the cooling system.

Although these works, and many others related to this issue, are relevant and extremely important when you wish to design and develop systems for irradiation of solid targets, which operate under conditions of high flux of particles and high performance, none of them addresses the effects of thermal transients and gradients of temperature.

In this work, a model was developed that takes into account not only the state of the system, but mainly the transient to the start of irradiation, when the stress suffered by the target material is great.

3. PRESENTATION OF THE PROBLEM

Figure 1 illustrates the geometry employed in this work. It shows an insert cylindrical shape of radius "a" and thickness "e".

This chip is irradiated by a beam of protons, represented by the function space q(r) which, in most practical cases, can be written in the form of a truncated Gaussian, but this work will be considered as a constant function.

Let T(r,z,t) the temperature of the chip in a generic point of coordinates (r,z) at a time "t". As the interest is in the gradients of temperature, one can write:

$$\theta(r,z,t)=T(r,z,t)-T_0 \tag{3}$$

For the problem, the equation of diffusion of heat is written as follows [6]:

$$\nabla^2\theta=\frac{1}{\alpha}\frac{\partial\theta}{\partial t} \quad \text{com} \quad \alpha=\frac{K}{\rho.c} \tag{4}$$

Where "ρ" is the density of the irradiated target, "c" the specific heat and "K" the material thermal conductivity.

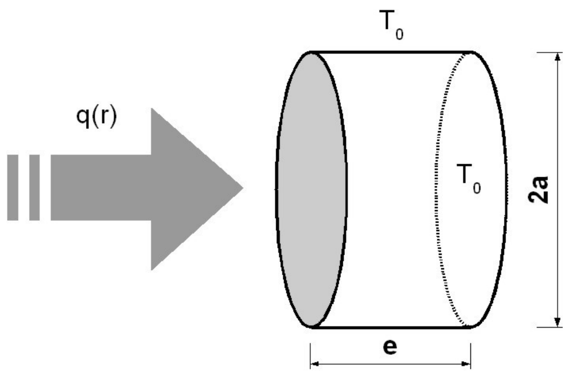

Figure 1. Solid target in the form of cylindrical pellet of radius "a" and thickness "e" bombarded by the beam of protons represented by q (r).

The initial and boundary conditions for this problem are given by the following expressions:

$$T(r,z,0)=T_0 \Rightarrow \theta(r,z,0)=0 \quad (5)$$

$$\begin{aligned} -K\frac{\partial \theta}{\partial z} &= q(r) \\ \theta(\mathrm{r,e,t}) &= 0 \end{aligned} \quad (6)$$

Set the initial and boundary conditions of the problem, the solutions that makes it physically possible, and then writes up the sought solution in the form of a superposition of two functions:

$$\theta(\mathrm{r,z,t})=\psi(\mathrm{r,z,t})+\varphi(\mathrm{r,z}) \quad (7)$$

The first term on the left side of this expression represents the thermal transient while the second term is the stationary state.

Figure 2 illustrates how the functions form, given in (7), should be.

3.1. Solution for the Stationary State

On balance, it is expected that $\theta(\mathrm{r,z,t}\rightarrow\infty)\approx\varphi(\mathrm{r,z})$, so that the boundary conditions for "r" and "z" are:

$$\begin{aligned} &\varphi(a,z)=0 \quad \mathrm{e} \quad \varphi(\mathrm{r,e})=0 \\ &\frac{\partial \varphi(0,z)}{\partial r}=0 \quad \mathrm{e} \quad \frac{\partial \varphi(\mathrm{r},0)}{\partial z}=-\frac{q(r)}{K} \end{aligned} \quad (8)$$

And the solution to the permanent regime is:

$$\varphi(r,z)=\sum_{n=1}^{\infty} a_n J_0(\lambda_n r).\sinh\left[\lambda_n(e-z)\right] \tag{9}$$

With:

$$a_n=\frac{\frac{2}{K}\int_0^a q(r).r.J_0(\lambda_n r)\,dr}{\lambda_n \cosh(\lambda_n e) a^2 J_1^2(\lambda_n a)} \tag{10}$$

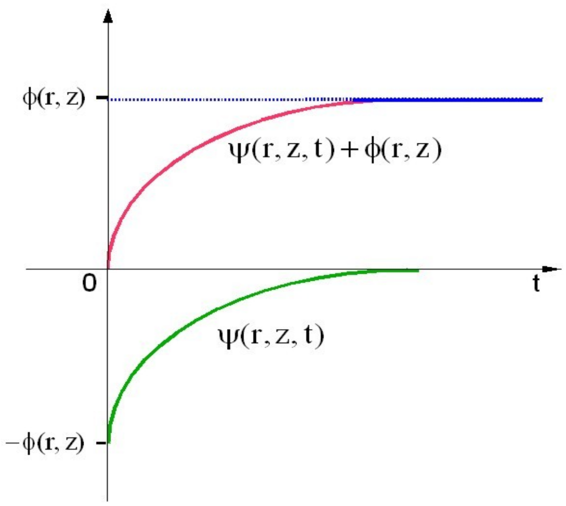

Figure 2. Linear superposition of solutions $\psi(\mathrm{r,z,t})e\,\varphi(\mathrm{r,z})$.

3.2. Solution for the Thermal Transient

The initial and boundary conditions for the transient heat equation are respectively:

$$\psi(r,z,0)=-\varphi(r,z) \tag{11}$$

$$\begin{gathered}\psi(a,z,t)=0 \text{ e } \psi(\mathrm{r,e,t})=0\\ \frac{\partial\psi(0,\mathrm{z,t})}{\partial r}=0 \text{ e } \frac{\partial\psi(\mathrm{r,0,t})}{\partial z}=0\end{gathered} \tag{12}$$

With this, the solution for $\psi(\mathrm{r,z,t})$ is:

$$\psi(r,z,t)=-\sum_{n=1}^{\infty}\sum_{k=1}^{\infty}\frac{2\mathrm{a}_k\beta_k\cosh(\beta_k e)}{e.(\mu_n^2+\beta_k^2)}\times e^{-\alpha\left(\mu_n^2+\beta_k^2\right)t}\cos(\mu_n z)J_0(\beta_k r) \tag{13}$$

Finally, the general solution for the target temperature at anywhere and at any time is given by:

$$T(r,z,t) = -\sum_{n=1}^{\infty}\sum_{k=1}^{\infty} \frac{2.a_k\beta_k \cosh(\beta_k e)}{e.(\mu_n^2+\beta_k^2)} \times e^{-\alpha\left(\mu_n^2+\beta_k^2\right)t} \cos(\mu_n z) J_0(\beta_k r) +$$
$$+\sum_{k=1}^{\infty} \frac{\frac{2}{K}\int_0^a q(r) r J_0(\beta_k r)\,dr}{\beta_k \cosh(\beta_k e) a^2 J_1^2(\beta_k a)} \times J_0(\beta_k r)\sinh[\beta_k(e-z)] + T_0 \qquad (14)$$

The constants of integration a_k e β_k are determined by the initial and the boundary conditions imposed on the $T(r,z,t)$, and the eingenvalues values μ_n are determined by:

$$\mu_n = \frac{(2\mathrm{n}-1)\pi}{2.e} \qquad (15)$$

4. RESULTS

Table 2 shows the specific heat, density and thermal conductivity of some materials used as target materials [7], which were used in the simulations.

In the calculations, were considered q_0 thermal power densities between 1.88 MW/m^2 to 150 MW/m^2, values that are commonly found in the irradiation of solid targets.

Table 2. Physical properties of some materials most used as targets in the Production of radioisotopes of medical interest.

Material of the Target	Specific Heat $(J/kg.^oC)\times 10^3$	Density $(kg/m^3)\times 10^3$	Thermal Conductivity $(W/m.^oC)$	$\alpha = K/\rho.c$ (m^2/s)x10^{-6}
Copper	0.38	8.96	398.00	116.89
Thallium	0.13	11.85	390.00	253.16
Tellurium	0.21	6.24	5.90	4.50
Nickel	0.44	8.80	91.00	23.50

Figure 3 shows the results for the simulation of copper irradiation with 24 MeV beam energy and a 0.5 mA current, which corresponds to a thermal power of 12,000 W (150 MW/m^2).

The copper target, irradiated with the highest power density, is representative with regard to the high beam current (0.5 mA), and the temperature recorded on the face of the target was around 500 °C, far below the value melting temperature of this metal, of 1,084 °C. This fact makes clear that the copper targets can be irradiated with a 1.0 mA current, resulting in a high yield for production of ^{62}Cu by route $^{63}Cu(p,2n)^{62}Cu$.

Figure 4 shows the results for nickel irradiation simulation with 18 MeV beam energy and a 0.3 mA current, corresponding to a 5,400 W (67.5 MW/m^2) thermal power.

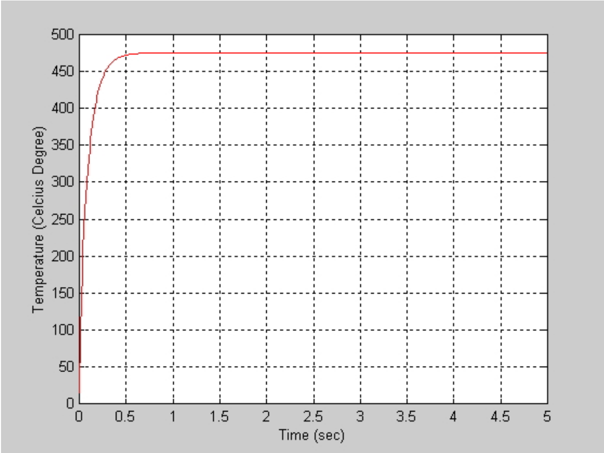

Figure 3. Thermal transient simulation for copper irradiated with 24 MeV proton beam and 0.5 mA current at the position z = 0.1 mm and r = 0.

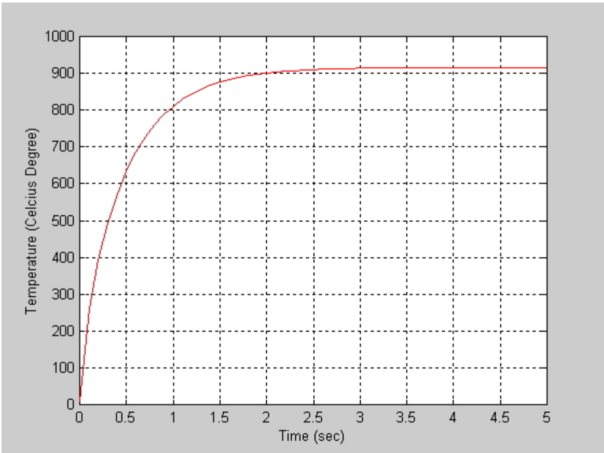

Figure 4.Thermal transient simulation for nickel irradiated with 18 MeV proton beam and a 0.3 mA current at the position z = 0.1 mm and r = 0.

The results for the nickel irradiation simulation are also very relevant, given the current interest in ^{64}Cu produced by route $^{64}Ni(p,n)^{64}Cu$.

The Nickel, presenting the greatest melting point among the studied materials (1,455 ° C), once used as target material may be irradiated with beam currents between 0.3 mA and 0.5 mA, not reaching the fusion temperature.

In thermal equilibrium and irradiated with current of 0.15 mA, it was found that the temperature of the nickel target surface was around 500 °C, and the transient thermal radiation with a current of 0.3 mA, the equilibrium was reached around 900 °C.

Figure 5 shows the results for the thallium irradiation simulation with 28 MeV beam energy and a 0.19 mA current, corresponding to a 5,320 W (≈ 66.5 MW/m^2) thermal power.

The results for thallium in accordance with the solution for the thermal diffusion profile in metals irradiated, the only case where experimental data are available. Although the metal of lowest melting point (304 ºC), has high thermal conductivity, only lower than copper's, and this can be irradiated with high beam currents.

For a current of 0.19 mA, the target surface temperature was around 200 °C, and near the middle of the target around 100 ºC. These values are consistent with those observed in the irradiation of thallium in IPEN.

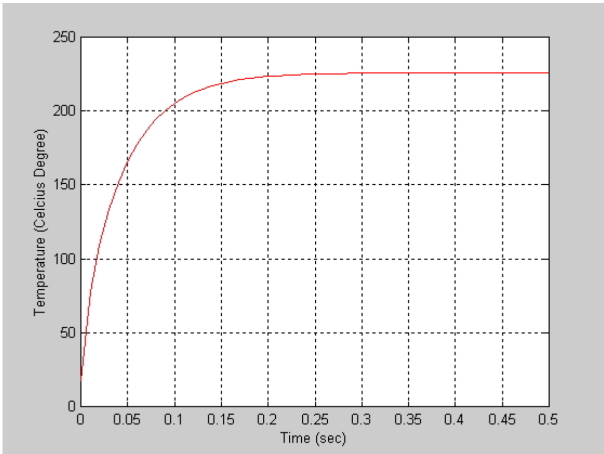

Figure 5. Thermal transient simulation for thallium irradiated with 28 MeV proton beam and a 0.19 mA current at the position z = 0.1 mm and r = 0.

Figure 6 shows the results for the tellurium irradiation simulation with beam energy of 12 MeV and a 0.125 mA, current, corresponding to a150 W (1.88 MW/m^2) thermal power.

Tellurium, being a semi-metal, has more serious limitations to the processes of irradiation with high beam currents. It has low melting point (449.5 °C) and low thermal conductivity, and therefore not used pure, but in the form of an oxide (TeO_2). This resolves the question of the melting point (~700 ºC) but does not improve the thermal conductivity factor. The results presented here are in complete agreement with those reported in the literature.

In all cases, the refrigeration system equilibrium temperature (T_0) was always maintained at 20 °C, and the calculations were all performed in a single point of coordinates (r = 0, z = 0.1 mm).

The energy chosen for each case are those that maximize the cross section of nuclear reaction to obtain the interest radioisotope, and the beam currents were chosen in order to maintain the temperature of each target below the melting or sublimation point to ensure the integrity of them.

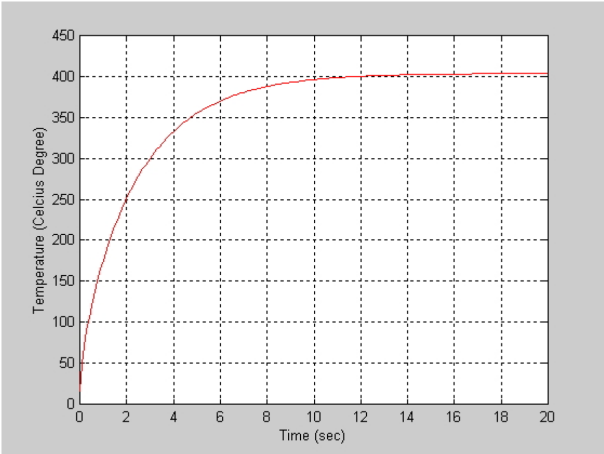

Figure 6. Thermal transient simulation for tellurium irradiated with 12 MeV proton beam and a 0.125 mA current at the position z = 0.1 mm and r = 0.

5. CONCLUSIONS

With the results obtained in the performed simulations, it was clear the importance of making a careful analysis of all parameters involved in the irradiation of solid targets, especially with regard to cooling.

Although the materials used in the simulations have physical and chemical characteristics very different in all cases it was observed that the time constant of the end of the transient solution of the diffusion equation dependent on the time showed the order of 10^7 s^{-1}. This shows that the deposit of energy on the target surface occurs in a few seconds.

It was evident that the temperatures near the target face, the most exposed to heat stress, remained well below the material melting or sublimation temperature, which certainly ensures the physical integrity of them.

With an improvement in engineering from the irradiation station and the implementation of a more efficient cooling system, one can irradiate this metal with proton beam current up to 0.45 mA.

Finally, we can conclude that the solution to the thermal diffusion model in irradiated targets can and should be used in the present and future of irradiation engineering systems, when seeking high performance and high efficiency. This will lead to an optimization routine in the production of ^{67}Ga and ^{201}Tl, reducing the time of irradiation, therefore the cyclotron life, and also in future irradiation to the ^{64}Cu and ^{124}I production.

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REFERENCES

1. International Atomic Energy Agency – IAEA-DCRP/2006, "Directory of Cyclotrons used for Radionuclide Production in Member States – 2006 updates", October 2006.
2. F. M. Nortier, N. R. Stevenson and W. Z. Gelbart, "*Investigation of the thermal perfomance of solid targets for radioisotope production*", Nuclear Instruments and methods in Physics Research A, 355 (1995) 236-241.
3. W. L. Talbert, D. M. Drake, H.-H. Hsu and M. T. Wilson, "*Design aspects of high-power targets*", Nuclear Instruments and methods in Physics Research A, 521 (2004) 136-142.
4. R. A. Pavan, W. Z. Gelbart and S. K. Zeisler, "*Thermal modeling of high-current solid targets*", Journal of Radioanalytical and Nuclear Chemistry, 257(1) (2003) 203-206.
5. J. J. Čonor, Ž. Stevanović, M. Rajčević and D. Košutić, "*Modeling of thermal properties of a TeO_2 target for radiodine production*", Nuclear Instruments and methods in Physics Research A, 521 (2004) 161-170.
6. "Conduction Heat Transfer", Vedat S. Arpaci, Addison-Wesley Publishing Company, ISBN 0-201-00359-7.
7. Handbook of Materials Science, Vol. I – General Properties, Charles T. Lynch, CRC Press Inc., Boca Raton, Florida, 1988.