JOURNALOF

Ceramic Processing Research

Effects of Al₂O₃ addition on nanocrystal formation and crystallization kinetics in (1-*x*)Li₂B₄O₇-*x*Al₂O₃ glasses

Hyun Woo Choi^a, Su Jae Kim^b, Hang Yang^b, Yong Suk Yang^b, Young Hoon Rim^c and Chae Ryong Cho^{a,b,*}

^aDepartment of Nanoenergy Engineering, Pusan National University, Busan 46241, Korea

^bDepartment of Nano Fusion Technology, Pusan National University, Busan 46241, Korea

^cCollege of General Education, Semyung University, Jecheon, Chungcheongbuk-do 27136, Korea

We investigated the effects of Al_2O_3 addition on $(1-x)Li_2B_4O_7-xAl_2O_3$ (LBAO; x = 0, 0.005, 0.01, 0.05, 0.07, and 0.1) glasses. The glasses were synthesized by a conventional melt-quench method. Structural transformations of the LBAO glasses were assessed via X-ray diffraction analysis. Estimations of ΔT , $K_{CS} = (T_c - T_g)/(T_m - T_c)$, activation energy, and the Avrami parameter were performed using differential thermal analysis and differential scanning calorimetry. An interpretation of non-isothermal kinetics of the crystallization process is presented using the modified Ozawa equation. The activation energy E increased from 3.3 to 3.5 eV for the LBAO (x < 0.01) glasses whereas those of the LBAO (x > 0.05) glasses slightly increased from 3.75 to 4.05 eV. The exponent *n* was estimated to be 3.9 ± 0.1 for the LBAO (x < 0.01) glasses and 3.2 ± 0.02 for the LBAO (x > 0.05) glasses. Microstructural characterization of the glassy and crystalline phases using atomic force microscopy was investigated. The effects of Al_2O_3 on the LBAO glasses include a decreased nucleation rate in the crystallization process and a significantly reduced crystal size.

Key words: Thermal analysis, Nanocrystalline materials, Li₂B₄O₇-Al₂O₃ glass, Growth kinetics, Crystallization.

Introduction

Nowadays, there is growing interest in the development of novel materials to satisfy specific target requirements. It is well known that nanomaterials usually exhibit properties different than those of the corresponding bulk materials. The crystallization processing of glasses has become increasingly important as a novel technique for the development of novel functional glass/crystal hybrid materials as well as an approach for gaining a detailed understanding of the glass structures.

The lithium tetraborate ($\text{Li}_2\text{B}_4\text{O}_7$; LBO) crystal is a material commonly used in surface acoustic-wave substrates and in nonlinear devices for frequency conversion in the ultraviolet region as well as a piezoelectric actuator [1-3]. The simple fabrication method of the LBO glass facilitates highly efficient production of complex-shaped and multi-sized forms. In addition, the formation of microcrystals from glass can be observed by optical microscopy, as the LBO glass and crystal are colorless and transparent.

In a recent review paper, Komatsu introduced the basic concept of the oxide glass structure in relation to crystallization, the design and control of crystal morphologies, and glass-ceramic properties along with some relative examples [4]. He emphasized that the glass-forming tendency and the glass-forming region can provide valuable information on the nanoscale heterogeneous structure.

Varma's research group [5, 6] has succeeded in crystallization of various optically active and ferroelectric nanocrystals such as $BaBi_2Nb_2O_9$ and Bi_2WO_6 in $Li_2B_4O_7$ -based glasses. Several other researchers have investigated the effects of the addition of Al_2O_3 in terms of improving the stability of SiO₂–BaO–TiO₂ glasses [7,8]. However, to the best of our knowledge, there has been no study on the addition and crystallization behavior of Al_2O_3 in $Li_2B_4O_7$ -based glasses.

In this study, we investigated the structural and thermal properties of $(1-x)Li_2B_4O_7$ - xAl_2O_3 (LBAO; x = 0, 0.005, 0.01, 0.05, 0.07, and 0.1) glasses to understand the effects of Al_2O_3 on the formation of nanocrystals and the crystallization kinetics in LBAO glass systems. Moreover, the chemical bonding states of the elements in the amorphous and annealed samples were studied.

Experimental

The $(1-x)Li_2B_4O_7-xAl_2O_3$ (LBAO, x = 0.005, 0.01, 0.05, 0.07, 0.1) glasses were fabricated by a conventional melt-quench method. Li_2CO_3 (0.01 mol, 99.7%, Sigma-Aldrich) and B_2O_3 (0.02 mol, 99%, Sigma-Aldrich) were used as sources of Li and B, respectively. After pyrolysis of the product at 850 °C, a

^{*}Corresponding author:

Tel:+82-51-510-6114

Fax: +82-51-514-2358 E-mail: crcho@pusan.ac.kr

powder precursor, Li₂B₄O₇, was obtained. Al₂O₃ (10 µm, 99.7%, Aldrich) and Li₂B₄O₇ powders were mixed well for 2 hrs to improve the homogeneity. In this method, the melt was quenched by pouring it onto a copper plate and immediately pressing it with a second plate to maximize the cooling rate. All glasses were synthesized to a thickness of approximately 1 mm, and were transparent. The amorphous natures of the LBAO glasses were confirmed by X-ray diffraction (XRD; Miniflex II, Rigaku, Japan) analysis with Cu K_{α} radiation ($\lambda = 1.5406$ Å) and differential scanning calorimetry (DSC; DSC3100s, MAC Science, Japan). The chemical bonding states of the samples were analyzed by X-ray photoelectron spectroscopy (XPS, K-ALPHA⁺, Thermo Fisher Scientific, USA) with monochromatic Al-K_a radiation (1486.6 eV) in the range 0-1200 eV. The binding energy in the spectra was calibrated to carbon (C 1s, 284.6 eV). The nonisothermal crystallization kinetics of the LBAO glasses were analyzed using DSC under a pure argon atmosphere. The sample (5 mg) was encapsulated in a platinum pan and was heated at various rates for nonisothermal experiments. Continuous heating experiments were performed with programs beginning at room temperature and heating to 600 °C at scan rates of 2, 5, 7, and 10 °C/min. An empty pan was used to obtain a baseline subtraction for each measurement. Thermogravimetric differential thermal analyses (TG-DTA; TGand DTA2020s, MAC Science, Japan) were performed to measure the thermal change during the transition of the melt. The surface morphologies were analyzed using atomic force microscopy (AFM; Park XE7, Park scientific, Korea). To estimate the average grain sizes, we used the Scherrer equation with the XRD data, and AFM images using the program (Park SmartScan) of the AFM control computer.

Results and Discussion

Fig. 1 shows XRD patterns of the (a) as-quenched (1x)Li₂B₄O₇-xAl₂O₃ (LBAO, x = 0.005, 0.01, 0.05, 0.07,0.1) glasses and (b) glasses heat-treated at 700 °C for 10 min. The as-quenched samples exhibited the typical broad peaks attributed to the short-range order of the glass state. For the LBAO (x = 0.005 and 0.01) glasses treated at 700 °C for 10 min, peaks attributed to tetragonal crystals of $Li_2B_4O_7$ were observed. Furthermore increase in the concentration of Al₂O₃, as the LBAO (x > 0.05) samples, led to the formation of a major tetragonal phase with a minor monoclinic phase of Li₂AlB₅O₁₀, indicating the coexistence of tetragonal and monoclinic phases. Fig. 1(b) shows the crystallization structure of the samples and its dependence on the proportion of Al₂O₃ in the composition. Based on the results, it was suggested that the solubility limit of Al₂O₃ in LBO is about 7%. The apparent mixed phase of the Li₂AlB₅O₁₀ crystal structure appeared at 10%



Fig. 1. (a) XRD patterns of the as-quenched $(1-x)Li_2B_4O_7-xAl_2O_3$ glasses at room temperature and (b) of glasses heat-treated at 700 °C for 10 min. The symbols σ and δ represent the positions of the peaks of $Li_2B_4O_7$ and $Li_2AlB_5O_{10}$, respectively. The ICSD patterns of the tetragonal (no. 18996; symbol σ) and monoclinic (no. 64672; symbol δ) structures are included.

 Al_2O_3 content. Differences in the intensities of the peaks at 33.6 ° [LBO (213)] and 34.6 ° [LBO (312)] were also observed with varying Al_2O_3 composition. To clarify this, we performed XRD intensity simulations with variations in the atomic occupancy.

Fig. 2 shows the simulation result and atomic site of the LBO crystal. The LBO crystal had a tetragonal structure with space group I41cd, containing 1 Li, 2 B and 4 O sites [9]. As shown in Fig. 2(a), the simulation was carried out with the condition of 5% of Al at the B1 and B2 sites, and 20% vacancy at the O1, O2, O3, and O4 sites. The LBO (213) peak was more intense than the LBO (312) peak when the O3 site was vacant. In contrast, the LBO (312) peak had greater intensity than the LBO (213) peak when the O1 site was vacant. Based on the experimental and simulation results, it was determined that the crystal phase had a vacancy at the O3 site at low Al_2O_3 concentrations, but that this vacancy was occupied and the O1 site was left vacant with increasing Al₂O₃ concentration. The reason underlying the creation of this O1 site vacancy is still



Fig. 2. (a) simulated XRD pattern of $Li_2B_4O_7$ crystal with various defect or substitution conditions, and (b) model structure of $Li_2B_4O_7$ tetragonal crystal.



Fig. 3. XRD patterns of $(1-x)Li_2B_4O_7-xAl_2O_3$ (x = 0.07) glass recorded at room temperature for the samples quenched at the given temperatures after annealing at a heating rate of 10 °C /min.

under investigation.

Fig. 3 shows XRD patterns of the as-quenched LBAO (x = 0.07) glasses (bulk state) heat-treated at different temperatures. The XRD patterns were recorded at room temperature, and the samples were obtained by annealing the LBAO (x = 0.07) glass by heating it to a given temperature in an electric furnace at a heating rate of 10 °C/min. The XRD pattern of the as-quenched sample shows the typical broad diffraction shape from the short order of the glass state. The sample quenched at 590 °C exhibited a single phase of tetragonal Li₂B₄O₇ crystal in the XRD pattern. For the glass treated at 700 °C, an additional peak at $2\theta = 17.02^{\circ}$ was observed, which was attributed to the formation of monoclinic Li₂AlB₅O₁₀, which demonstrates coexisting tetragonal and monoclinic phases. The lattice parameters obtained from the data were: a = b = 9.479 Å, c = 10.286Å, $\alpha = \beta = \gamma = 90^{\circ}$ for the tetragonal Li₂B₄O₇ (ICSD no. 18996), and a = c = 7.04 Å, b = 14.94 Å, $\alpha = \gamma = 90^{\circ}$, $\beta =$ 90.7° for the monoclinic $Li_2AlB_5O_{10}$ (ICSD no. 64672). DTA and DSC were performed for all quenched samples to understand the effect of Al₂O₃ on the crystallization process of the LBAO system. The temperature parameters are herein denoted follows: T_g , glass transition temperature;



Fig. 4. (a) XPS wide-scan spectra of $(1-x)Li_2B_4O_7$ -xAl₂O₃ (x = 0.01-0.1) glasses. (b-d) XPS narrow-scan spectra of Li 1s, Al 2p, O 1s, and B 1s peaks, respectively for the glass samples.



Fig. 5. (a) XPS wide-scan spectra of $(1-x)Li_2B_4O_7-xAl_2O_3$ (x = 0.01-0.1) glasses annealed at 700 °C. (b-d) XPS narrow-scan spectra of Li 1s, Al 2p, O 1s, and B 1s peaks, respectively for the annealed samples.

 T_c , crystallization temperature; T_m , melting temperature; ΔT , stability against crystallization; T_p , maximum exothermic temperature:

Fig. 4 displays the existence of Li, Al, O, and B in $(1-x)Li_2B_4O_7$ - xAl_2O_3 glasses at different compositions of x. To obtain detailed information about chemical bonding states, wide and narrow XPS scans were performed for each element, as shown in Figs. 4(a) and 4(b-d), respectively. With increasing Al content, the peak area of Al increased due to the uniform distribution of the added element. The position and shape of each of the Li 1s, Al 2p, O 1s, and B 1s peaks were very similar between all samples. Neither a specific peak behavior nor shift of chemical bonding state were observed for any element. The O 1s binding energy of 531.7 eV confirms the O_2^- state of oxygen in the materials, which bound to the boron, lithium, or aluminum in the lattice.

The chemical bonding states of the elements in



Fig. 6. DTA curves of the $(1-x)Li_2B_4O_7-xAl_2O_3$ glasses, measured at a heating rate of 10 °C/min. T_g , T_c , T_p , and T_m denote the glass transition temperature, crystallization temperature, maximum exothermic temperature, and melting temperature, respectively.



Fig. 7. First exothermic peaks in the DSC thermographs during crystallizations of the $(1-x)Li_2B_4O_7-xAl_2O_3$ (x = 0.07) glass at heating rates of 2, 5, 7, and 10 °C/min. The inset shows the specific temperatures of T_g , T_c , and T_p at the heating rate of 7 °C/min.

samples annealed at 700 °C are shown in Fig. 5. The main structural pattern in the $Li_2B_4O_7$ unit cell was the [B₄O₉]⁶⁻ complex, which consists of bridging BO₃ and BO₄ with Li⁺ ions distributed throughout. In contrast, the Li₂AlB₅O₁₀ unit cell was characterized by an AlO₄ tetrahedron and a three-dimensional network of $[B_5O_{10}]^{5}$ groups, which consist of two planar hexagonal B-O rings oriented perpendicularly to each other and linked by a tetra-coordinated B atom, and tetra-coordinated Li⁺ cations [10-12]. At higher annealing temperatures, the Li⁺ could easily evaporate because of the formation of defects during the crystallization of the amorphous structure. With increasing Al₂O₃ content, the bond strength of Li⁺ was also weakened, further facilitating the evaporation of Li⁺. This behavior corresponds to the observations of the Li 1s peak of the XPS spectrum of LBAO (x = 0.1) shown in Fig. 5(b). Moreover, the peak behaviors of XPS Li 1s, Al 2p, O 1s, and B 1s

Table 1. T_g , T_c , T_m , ΔT , and	T_p at various compositions of x .
	Concentration (<i>x</i>)

	Concentration (x)						
Temp. (° <i>C</i>)	0	0.005	0.01	0.05	0.07	0.1	
Tg	481	480	480	476	477	475	
T _c	530.6	535	536	550	556	560	
$ \Delta T \\ (=T_c - T_g) $	49.6	55	56	74	79	85	
T _{p1}	543	551	555	567	570	578	
T_{p2}				676	670	665	
T_{m1}	909	908	900	893	887	883	
T _{m2}				785	793	795	

due to the formation of $Li_2AlB_5O_{10}$ were appeared, as similar to that of XRD shown in Fig. 1.

Fig. 6 shows DTA thermograms of the LBAO glasses with different Al_2O_3 contents (x) measured at a heating rate of 10 °C/min. With addition of Al₂O₃, the first exothermic peak T_{pl} gradually shifted to higher temperatures, but the second exothermic peak T_{p2} gradually shifted to lower temperatures. On the contrary, the first endothermic melting tesmperature T_{m1} gradually decreased, while the second endothermic melting temperature T_{m2} gradually increased, with increasing Al₂O₃ content. In order to discuss the relationships between the non-isothermal crystallization and glass stability against devitrification upon heating, the various temperatures obtained from the DTA measurements are summarized in Table 1. Considering the first endothermic peaks of the LBAO glasses, as shown in Fig. 6, the glass stability against devitrification upon heating, KGS (= $(T_c - T_g)/(T_m - T_c)$), increased with further addition of Al₂O₃ to in the LBAO glasses. This result implies that the Al₂O₃-free glass of the LBAO system crystallizes more readily than the LBAO glass with Al_2O_3 added [13].

Fig. 7 shows several non-isothermal DSC curves for the LBAO glass (x = 0.07) measured at heating rates of 2-10 °C/min between 400 and 600 °C. The peak corresponds to the first crystallization peak, which indicates a single tetragonal Li₂B₄O₇ crystal phase in the temperature range of 400 to 600 °C. The inset of Fig. 7 shows the extrapolation used to determine the values of T_g and T_c at the heating rate of 7 °C/min. The normalized exothermic peak in Fig. 7 shifted toward higher temperatures and increased in intensity with increasing heating rate. The normalized peak per unit mass is a useful parameter to evaluate the parameters of crystallization, as the exothermic peak area is proportional to the sample mass. The transformed volume fraction, x(t) of the crystal at each heating rate was obtained as a function of temperature from the area under the corresponding DSC curve. Integration of the DSC curve over the temperature range then yielded the evolution of the peak area with time.



Fig. 8. (a) Activation energy as a function of the crystallization fraction *x*, determined by the modified Ozawa model. The inset shows the process used to calculate the activation energy from the slope of the logarithmic function plotted against 1/T. (b) Avrami exponent *n* calculated using the modified Ozawa model. Inset: the straight lines represent the linear regressions of ln[-ln(1-*x*)] against -ln α at different temperatures, yielding linear dependences with the Avrami exponent n = 3.2.

The typical theoretical bases describing the nonisothermal crystallization of glasses are introduced by the Johnson-Mehl-Avrami-Kolmogorov (JMAK) and modified Ozawa equations [14-17].

The modified Ozawa equation for linear heating [18,19] is expressed as:

$$\ln[-\ln(1-x)] = -n\ln\alpha - 1.052m(E/kT) + \text{const.}$$
(1)

where x is the crystallized volume fraction, n is the Avrami exponent, α is the heating rate, *m* is the dimension of crystal growth, E is the activation energy of crystallization in the glass, R is the ideal gas constant, and T is the temperature. The plot of $\ln[-\ln(1$ x)] as a function of 1/T or $\ln \alpha$ yields a straight line, whose slope reveals the activation energy or Avrami exponent. Fig. 8(a) shows that the measured activation energies increased from 3.3 to 3.5 eV for the LBAO (x < 0.01) glasses, while those for the LBAO (x > 0.05) glasses increased slightly from 3.75 to 4.05 eV. Fig. 8(b) shows that the Avrami exponent *n* was 3.9 ± 0.1 for the LBAO (x < 0.01) glasses and 3.2 ± 0.02 for the LBAO (x > 0.05) glasses. The inset of Fig. 8(a) shows the activation energies obtained for the LBAO (x =0.07) sample at various heating rates and temperatures. From the slope of $\ln[-\ln(1-x)]$ as a function of $\ln\alpha$ in the inset of Fig. 8(b), we obtained the Avrami exponent n, which depends on the nucleation and growth processes. For example, an n of 3.2 was obtained for the LBAO (x = 0.07) sample.

The value of the growth dimension was m = 3, as obtained from the AFM images shown in Fig. 9, indicating that the corresponding mechanism for the LBAO (x < 0.01) glasses was interface-controlled growth with a constant nucleation rate. This result is consistent with previous isothermal measurements made by Kim et al. [20]. For the LBAO (x > 0.05) glasses, the mechanism



Fig. 9. AFM images of the $(1-x)Li_2B_4O_7-xAl_2O_3$ (x = 0.07) glass acquired at room temperature after annealing the glass samples at 560, 565, 570, 575, and 590 °C.

Table 2. Average sizes of the crystal grains for the LBAO (x = 0 and 0.07) glasses obtained from the XRD data and AFM morphologies at various temperatures. The average crystal grain sizes are calculated from the XRD data using the Debye-Scherrer equation.

For $x=0$ (Li ₂ B ₄ O ₇)							
	560 °C	565 °C	570 °C	575 °C	590 °C		
XRD	32 ± 2 nm	$168 \pm 2 \text{ nm}$	$95\pm4\ nm$	$132\pm3\ nm$	$165\pm11 \text{ nm}$		
AFM	35 ± 2 nm	$168 \pm 3 \text{ nm}$	$102 \pm 2 \text{ nm}$	$158\pm3\ nm$	$205\pm 6 \ nm$		
For $x=0.07$ (Li ₂ B ₄ O ₇ + Al ₂ O ₃)							
	560 °C	565 °C	570 °C	575 °C	590 °C		
XRD	$28 \pm 3 \text{ nm}$	$n38 \pm 2 \text{ nm}$	$55\pm\ 2\ nm$	$100\pm5~\text{nm}$	$125\pm7nm$		
AFM	30 ± 2 nm	$142 \pm 3 \text{ nm}$	$63\pm\ 3\ nm$	$105\pm2~\text{nm}$	160 ± 3 nm		

was interface-controlled growth with a decreasing nucleation rate. Therefore, one effect of Al2O3 in the LBAO glasses was a decreased nucleation rate in the crystallization process. In addition, the activation energies slightly increased with increased concentration of Al₂O₃ in the LBAO glasses. In order to understand the formation of nanocrystals from glass upon heating, we analyzed the XRD patterns and morphologies using the Scherrer equation and AFM images. The Scherrer equation is: $L = SF\lambda/\Gamma \cos\theta$, where L is the grain size, SF = 0.94 is the shape factor, λ is the X-ray wavelength of 1.5406 Å, Γ is the full width at half maximum of the Bragg peak, and θ is the peak position. Using an extended view of the XRD patterns for the Bragg (112) diffraction, we obtained the crystal grain sizes. AFM images of the fractured surfaces are shown in Fig. 9. The shapes of the crystallites were rock-like, while the dimensionality of their growth was m = 3.

Table 2 summarizes the average sizes of the crystal grains for the LBAO (x = 0 and 0.07) glasses obtained from XRD data and AFM morphologies obtained at various temperatures. The AFM analysis confirms the sizes obtained from the XRD measurements, even though the sizes obtained from the AFM images were slightly larger than those obtained via XRD. The formation of nanocrystals also occurred in the LBAO glasses, and the crystal sizes in the LBAO glasses continuously increased with increasing temperature. Compared to the Al₂O₃-free glass of the LBAO system, the crystal size significantly decreased with the addition of Al₂O₃ as in the LBAO (x = 0.07) glass, confirming that Al₂O₃ can suppress the crystallization of Li₂B₄O₇.

Conclusions

The addition of Al_2O_3 improved the stability against crystallization upon heating of the $(1-x)Li_2B_4O_7-xAl_2O_3$ glass system by changing the crystallization mechanism.

The activation energies slightly increased with increased concentration of Al_2O_3 in the LBAO glasses. For the LBAO (x < 0.01) glasses, the mechanism was interfacecontrolled growth with a constant nucleation rate whereas that for the LBAO (x > 0.05) glasses was interface-controlled growth with a decreasing nucleation rate. Therefore, the main effect of Al_2O_3 in the LBAO glasses was incurring a decrease in the nucleation rate in the crystallization process. The crystal size significantly decreased upon Al_2O_3 addition to the LBAO glass, confirming that Al_2O_3 suppressed the crystallization of the Li₂B₄O₇ glass.

Acknowledgements

This study was financially supported by the "2017 Post-Doctoral Development Program" of Pusan National University.

References

- 1. R. Komatsu, T. Sugawara, and K, Sassa, Appl. Phys. Lett. 70 (1997) 3492-3494.
- J.H. Cho, N.J. Bang, and S.H. Kim, J. Korean Phys. Soc. 29 (1996) 8555-8557.
- S.J. Kim, J.E. Kim, and Y.H. Rim, Solid State Commun. 131 (2004) 129-133.
- 4. T. Komatsu, J. Non-Cryst. Solids 428 (2015) 156-175.
- 5. G.S. Murugan, K.B.R. Varma, J. Non-Cryst. Solids 279 (2001) 1-13.
- C. Karthik, K.B.R. Varma, J. Non-Cryst. Solids 353 (2007) 1307-1310.
- X. Su, M. Tomozawa, J.K. Nelson, and D.B. Chrisey, J. Mater. Sci. Mater. EI. 24 (2013) 2135.
- J. Chen, Y. Zhang, and C. Deng, J. Am. Ceram. Soc. 92 (2009) 1350-1353.
- S.J. Kim, W.K. Kim, Y.C. Cho, S. Park, I.K. Jeong, Y.S. Yang, Y. Kuroiwa, C. Moriyoshi, H. Tanaka, M. Takata, and S.Y. Jeong, Curr. Appl. Phys. 11 (2011) 649-652.
- M. He, H. Li, and X. Chen, Acta. Crystallogr. C. 57 (2001) 1010-1011.
- M.M. Islam, V.V. Maslyuk, and T. Bredow, J. Phys. Chem. B. 109 (2005) 13597-13604.
- 12. T.D. Kelly, J.C. Petrosky, and J.W. McClory, Front. Phys. 2 (2014) 31.
- Avramov, I.E.D. Zanotto, and M.O. Prado, J. Non-Cryst. Solids 320 (2003) 9-20.
- 14. G. Ruitenberg, E. Woldt, and A.K. Petford-Long, Thermochim. Acta 278 (2001) 97.
- 15. M. Avrami, J. Chem. Phys. 7 (1939) 1103.
- 16. M. Avrami, J. Chem. Phys. 8 (1940) 212.
- 17. M. Avrami, J. Chem. Phys. 9 (1941) 177.
- K. Matusita, T. Komatsu, and R. Yokota, J. Mater. Sci. 19 (1984) 291-296.
- J.W. Christian, in "The Theory of Transformation in Metals and Alloys", (Pergamon Press, Oxford, 1975) p. 1.
- 20. S.J. Kim, J.E. Kim, and Y.S. Yang. Solid State Commun. 122 (2002) 135-138.