

Solid State Green Synthesis and Properties on Paracetamol-Phenobarbital Binary Drug System

H. Shekhar* and Raj Laxmi

Department of Chemistry, V.K.S. University, Ara, Bihar, India.

*Corresponding author: hshe2503@rediffmail.com

Abstract: Molecular interaction between nonsteroidal anti-inflammatory drug (NSAID)/analgesic/antipyretic drug and antiepileptic drug was studied taking views effectiveness and enhanced pharmaceutical properties. The objective of the present communication is to highlight thermodynamic and interfacial investigation of paracetamol (PCM) and phenobarbital (PB) binary drugs. The value of integral excess Gibbs energy gE in the PCM-PB system shows the positive deviation from ideal behaviour. It refers stronger association between the molecules of same drug during formation of binary mix. The negative value of Gibbs free energy of mixing (ΔG_M) suggests the mixing of parent drug in eutectic and non eutectic binary drug is spontaneous. The interfacial property of binary drugs has been determined by the help of interfacial energy (σ) and roughness parameter (α). The size of critical nucleus of binary drug at different undercoolings was in nano size is substantiated its great future for pharmaceutical industries.

Keywords: Binary drug, Critical radius, Excess and mixing thermodynamic function, Interfacial energy.

I. INTRODUCTION

The current development for Pharmaceutical drugs dosage tenders the pre-formulation designing [1-4] new drugs. The hydrophilic excipient strengthens the solubility of less soluble drugs and promotes the bioavailability and the therapeutic efficacy of drugs. Further, the binary product of excipient and drug would get sublimated quality and having high formulation properties such as the solubility, odor and taste of the drug. The past studies on drug association has not shown better performance with compare to generic drugs (e.g. powders, tablets and capsules). Paracetamol (PCM) belongs to NSAID (Non-Steroidal Anti-inflammatory Drugs) drugs [5-8] family which are used as pain and fever reliever. Past researches on binary drug products [9-11] prepared by PCM with Ascorbic acid and Citric acid had been highlighted and seen enhanced properties of paracetamol in powder form as well as in

solution. Keeping in view for minimizing overall side effect, Phenobarbital has been used in the treatment of epilepsy [12-16] and treat seizures in young children. It is also injected into a muscle and vein by mouth. The injection is given to treat status epilepticus. It is sometimes used to treat sleeplessness and anxiety. Its effect looks within five minutes when using injection and half an hour in case of orally taken and last upto two days. The present PCM-PB binary system has been taken a detailed investigation for thermodynamic and interfacial studies with concern to partial and integral thermodynamic quantities, interfacial energy, driving force of solidification, nanosize of critical radius and interfacial roughness.

II. EXPERIMENTAL DETAIL

Different ratios of drug (PCM) and excipient (PB) taken in test tube were heated and chilled in ice and made samples. The melting temperatures of solid dispersions of samples were determined using a precised apparatus. The enthalpy of fusion [17] of Paracetamol and Phenobarbital was determined by DTA method.

III. RESULTS AND DISCUSSION

A. Solid-Liquid Equilibrium Study

The melting point of PCM (M.P. - 168.7 °C) decreases on the addition of second component PB (M.P. - 174 °C). In Table I equilibrium data of solid dispersions of PCM-PB system have been mentioned and the binary drug system shows the formation of an eutectic (E) [18] and non-eutectics solid dispersion (A1-A14). The heat of fusion value for binary solid dispersions and eutectic has been presented in Table I. The activity coefficient and activity of components data for the present system has been determined [19] from the equation:

$$-\ln \chi_i \gamma_i = \frac{\Delta H_i}{R} \left(\frac{1}{T_e} - \frac{1}{T_i} \right) \quad (1)$$

where, ΔH_i is the heat of fusion of component i at melting point T_i . T_e is the melting temperature of solid dispersion. These values are very helpful in measuring the thermodynamic mixing and excess quantities.

B. Mixing and Excess Functions

The value of thermodynamic mixing functions; molar free energy (ΔG^M), molar entropy (ΔS^M) and molar enthalpy (ΔH^M) of the binary solid dispersions were determined by equations:

$$\Delta G^M = RT(\chi_{PCM} \ln a_{PCM} + \chi_{PB} \ln a_{PB}) \quad (2)$$

$$\Delta S^M = -R(\chi_{PCM} \ln \chi_{PCM} + \chi_{PB} \ln \chi_{PB}) \quad (3)$$

$$\Delta H^M = RT(\chi_{PCM} \ln \gamma_{PCM} + \chi_{PB} \ln \gamma_{PB}) \quad (4)$$

and partial molar free energy of thermodynamic mixing function, G_i^{-M} or mixing chemical potential (μ_i^{-M}) has been determined by the equation:

$$G_i^{-M} = \mu_i^{-M} = RT \ln a_i \quad (5)$$

The negative value [20] of molar free energy of mixing of solid dispersions shown in Table II suggests that the mixing in all cases takes place spontaneous. The integral excess thermodynamic functions in form of as integral excess integral free energy (g^E), excess integral entropy (s^E) and excess integral enthalpy (h^E) were determined using the equations:

$$g^E = RT (\chi_{PCM} \ln \gamma_{PCM} + \chi_{PB} \ln \gamma_{PB}) \quad (6)$$

$$s^E = -R \left(\chi_{PCM} \ln \gamma_{PCM} + \chi_{PB} \ln \gamma_{PB} + \chi_{PCM} T \frac{\delta \ln \gamma_{PCM}}{\delta T} + \chi_{PB} T \frac{\delta \ln \gamma_{PB}}{\delta T} \right) \quad (7)$$

$$h^E = RT \left(\chi_{PCM} \frac{\delta \ln \gamma_{PCM}}{\delta T} + \chi_{PB} \frac{\delta \ln \gamma_{PB}}{\delta T} \right) \quad (8)$$

The partial excess function (g_i^{-E}) or excess chemical potential (μ_i^{-E}) is determined by equation:

$$g_i^{-M} = \mu_i^{-M} = RT \ln \gamma_i \quad (9)$$

Table III forwards there is stronger interaction [21,22] between like molecules in entire binary dispersions.

C. The Solid-Liquid Interfacial Energy(σ)

The interfacial energy of binary solid dispersions calculated from melting enthalpy change [23] was determined by using Turnbull empirical relationship [24]:

$$\sigma = \frac{C \Delta H}{(N)^{1/3} (V_m)^{2/3}} \quad (10)$$

where, the coefficient C lies between 0.33 to 0.35 for nonmetallic system, N is the Avogadro's constant and V_m is molar volume. The value of σ for binary dispersions and parent components is mentioned in Table I which were in good agreement with the observed experimental values. In addition σ value was

evaluated using the equation:

$$\tau = r \Delta T = \frac{TV_m \sigma}{\Delta H} = \frac{\sigma}{\Delta S_v} \quad (11)$$

where, r is the radius grooves of interface and ΔT is the dispersion in equilibrium temperature. The value (Table I) of τ was also determined by the help of Gunduz and Hunt numerical method [25] for materials having grain boundary shape. The Gibbs-Thomson coefficient for PCM, PB and their solid dispersions are found in the range of 8.33 - 10.006 x 10⁻⁰⁶ Km. The value of interfacial grain boundary energy (σ_{gb}) was obtained by using equation [26]:

$$\sigma_{gb} = 2\sigma \cos \theta \quad (12)$$

θ has been found at zero contact angle. The value of σ_{gb} for pure components and all solid dispersions are reported in Table I.

D. The Driving Force of Nucleation (ΔG_v)

The effective entropy change per unit volume (ΔS_v) is given by:

$$\Delta S_v = \frac{\Delta H}{T} \cdot \frac{1}{V_m} \quad (13)$$

The value of ΔS_v are given in Table I decides the degree and orientation of ΔS_v . The stepwise/non-uniform surface leads at low driving force while continuous/uniform surface advances at sufficiently high driving force. The ΔG_v can be determined at different undercoolings (ΔT) by the equation [27] below.

$$\Delta G_v = \Delta S_v \Delta T \quad (14)$$

The value of ΔG_v for each solid dispersions and pure components has been reported in the Table IV.

E. The Critical Radius (r^*)

The critical size nucleus with radius r^* during nucleation was determined by the equation [28] mentioned below.

$$r^* = \frac{2\sigma}{\Delta G_v} = \frac{2\sigma T}{\Delta H_v \Delta T} \quad (15)$$

The value of r^* mentioned in Table V decreases with increase of undercooling and lies in nano range. The activation/critical free energy of nucleation (ΔG^*) is determined by the equation mentioned below [29].

$$\Delta G^* = \frac{16\pi\sigma^3}{3\Delta G_v^2} \quad (16)$$

The value of ΔG^* for all the products was given in Table VI.

F. Interface Morphology

During phase transformation the prediction of growth morphology of solid-liquid interface has been made from the value of the entropy of fusion and Jackson roughness parameter [30] as mentioned below.

$$\alpha = \xi \frac{\Delta H}{RT} = \xi \frac{\Delta S}{R} \quad (17)$$

The value of ξ lies $0 < \xi \leq 1$. The value of α given in Table I suggests the leading of the faceted growth [31] in the entire solid dispersions.

IV. CONCLUSION

The equilibrium phase diagram of PCM-PB system forms an eutectic solid dispersion. The negative value of ΔG^M and positive value of g^E for entire solid dispersions confirm the spontaneous mixing in all cases and there is stronger interaction between like molecules in binary dispersions. Faceted growth appearance has been predicted during the development of growth interface of binary product.

ACKNOWLEDGEMENT

We are thankful to the Head Department of Chemistry, V.K.S. University Ara, Bihar, India for providing research facilities.

REFERENCES

- [1] L. Kalantzi, C. Reppas, J. B. Dressman, G. L. Amidon, H. E. Junginger, K. K. Midha, V. P. Shah, S. A. Stavchansky, and D. M. Barends, "Biowaiver monographs for immediate release solid oral dosage forms acetaminophen (paracetamol)," *J Pharm Sci.*, vol. 95, no. 1, pp. 4-14, 2006.
- [2] J. L. Ford, "The current status of solid dispersions," *Pharm Acta Helv.*, vol. 61, no. 3, pp. 69-88, 1986.
- [3] H. Shekhar, and V. Kant, *Thermodynamics of Nicotinanamide Based Binary Drug System*, Lambert Acadmic Publishing, Germany, 2013.
- [4] J. Emese, and P. F. Nagymate, "The stability of vitamin C in different beverages," *Brit Food J.*, vol. 110, no. 3, pp. 296-309, 2008.
- [5] M. Alacqua, G. Trifir, E. Spina, S. Moretti, D. U. Tari, P. Bramanti, A. P. Caputi, and V. Arcoraci, "Newer and older antiepileptic drug use in southern Italy: A population-based study during the years 2003-2005," *Epilepsy Res.*, vol. 85, no. 1, pp. 107-113, 2009.
- [6] W. Mazurek, E. Mazureka, W. Winnicka, A. Czajkowska-Kosnik, U. Czyzewska, and W. Mityk, "Application of differentactions in tablets containing acetaminophen," *Acta Pol. Pharm.*, vol. 70, no. 5, pp. 787-793, 2012.
- [7] B. Tita, A. Fulias, G. Bandur, E. Marian, and D. Tita, "Compatibility study between ketoprofen and pharmaceutical excipients used in solid dosage forms," *J. Pharm. Biomed. Anal.*, vol. 56, no. 2, pp. 221-227, 2011.
- [8] U. Grundman, C. Womle, A. Biedler, S. Kreuer, M. Wrobel, and W. Wilhelm, "The efficacy of the non-opioid analgesics parecoxib, paracetamol and metamizol for postoperative pain relief after lumbar microdiscectomy," *Anesth. Analg.*, vol. 103, no. 1, pp. 217-222, 2006.
- [9] R. Sandulescu, S. Mirel, and R. Orian, "The development of spectrophotometric and electroanalytical methods for ascorbic acid effervescent dosage forms," *J. Pharm. Biomed. Anal.*, vol. 23, no. 1, pp. 77-87, 2000.
- [10] G. P. Bettinetti, P. Mura, A. Liguori, G. Bramanti, and F. Giordano, "Solubilization and interaction of naproxen with polyvinylpyrrolldone in aqueous solution and in the solid state," *Farmaco Prat.*, vol. 43, no. 11, pp. 331-343, 1988.
- [11] J. B. Marta, and Z. N. Jerzy, "Paracetamol: Mechanism of action, applications and safety concern," *Acta Polonica Pharmaceutica - Drug Research*, vol. 71, no. 1, pp. 11-23, 2014.
- [12] L. E. Aberg, M. Backman, E. Kirveskari, and P. Santavuori, "Epilepsy and antiepileptic drug therapy in juvenile neuronal ceroid lipofuscinosis," *Epilepsia*, vol. 41, no. 10, pp. 1296-1302, 2000.
- [13] P. Kwan, and M. J. Brodie, "Phenobarbital for the treatment of epilepsy in the 21st century: A critical review," *Epilepsia*, vol. 45, no. 9, pp. 1141-1149, 2004.
- [14] M. Bialer, "How did phenobarbital's chemical structure affect the development of subsequent antiepileptic drugs (AEDs)?," *Epilepsia*, vol. 53, no. Suppl. 8, pp. 3-11, 2012.
- [15] T. O. Crawford, W. G. Mitchell, I. S. Fishman, and S. R. Snodgrass, "Very high-dose phenobarbital for refractory status epilepticus in children," *Neurology*, vol. 38, no. 7, pp. 1035-1040, 1988.
- [16] K. I. Kwong, K. W. Tsui, S. P. Wu, A. Yung, E. Yav, F. Eva, C. K. Ma, S. Cherk, K. T. Liu, W. W. Cheng, and M. M. Yau, "Utilization of antiepileptic drugs in Hong Kong children," *Pediatr. Neurol.*, vol. 46, no. 5, pp. 281-286, 2012.
- [17] H. Shekhar, J. Kumar, and C. S. Saha, "Thermodynamic and interfacial studies of co-crystals of 8- hydroxyquinoline - Urea system," *Malaysian Journal of Chemistry*, vol. 15, no. 1, pp. 67-76, 2013.
- [18] H. Shekhar, and M. Kumar, "Phase diagram, thermodynamic stability and interfacial studies on solid dispersions of phenothiazine-acetanilide drug system," *Asian J. of Organic and Medicinal Chemistry*, vol. 1, pp. 26-32, 2016.
- [19] L. S. Darken, "Thermodynamics binary metallic solutions," *Trans. Met. Soc. AIMS*, vol. 239, pp. 80-89, 1987.
- [20] H. Shekhar, and V. Kant, "Thermodynamic studies on solid dispersions, of nicotinamide - Khellin drug system," *Mol. Cryst. Liq. Cryst.*, vol. 577, no. 1, pp. 103-115, 2013.

- [21] B. L. Sharma, S. Tandon, and S. Gupta, "Characteristics of the binary faceted eutectic: Benzoic Acid - Salicylic acid system," *Cryst. Res. Technol.*, vol. 44, no. 3, pp. 258-268, 2009.
- [22] M. Shamsuddin, S. B. Singh, and A. Nasar, "Thermodynamic investigations of liquid gallium-cadmium alloys," *Thermochimica Acta*, vol. 316, no. 1, pp. 11-19, 1998.
- [23] N. B. Singh, and M. E. Glicksman, "Determination of the mean solid-liquid interface energy of pivalic acid," *J. Cryst Growth*, vol. 98, no. 4, pp. 573-580, 1989.
- [24] D. Turnbull, "Formation of crystal nuclei in liquid metals," *J. Appl. Phys.*, vol. 21, pp. 1022-1027, 1950.
- [25] M. Gunduz, and J. D. Hunt, "Solid-liquid surface energy in the Al-Mg system," *Acta Metall.*, vol. 37, no. 7, pp. 1839-1845, 1989.
- [26] S. Akbulut, Y. Ocak, K. Keslioglu, and N. Marasli, "Determination of interfacial energies in the amino-methylpropanediol-neopentylglycol organic alloy," *Applied Surface Science*, vol. 255, pp. 3594-3599, 2009.
- [27] J. D. Hunt, and S. Z. Lu, *Hand Book of Crystal Growth*, D. T. J. Hurie, (ed.), Elsevier, Amsterdam, p. 112, 1994.
- [28] G. A. Chadwick, *Metallography of Phase Transformation*, Butterworths, London, p. 61, 1972.
- [29] W. R. Wilcox, "The relation between classical nucleation theory and the solubility of small particles," *Journal of Crystal Growth*, vol. 26, no. 1, pp. 153-154, 1974.
- [30] J. D. Hunt, and K. A. Jackson, "Binary eutectic solidification," *Trans. Metall. Soc. AIME*, vol. 236, pp. 843-852, 1966.
- [31] P. Gupta, T. Agrawal, S. S. Das, and N. B. Singh, "Phase equilibria and molecular interaction studies on (naphthols + vanillin) systems," *The Journal of Chemical Thermodynamics*, vol. 48, pp. 291-299, 2012.

TABLE I: PHASE COMPOSITION, MELTING TEMPERATURE, VALUES OF ENTROPY OF FUSION PER UNIT VOLUME (ΔS_V), HEAT OF FUSION (ΔH), INTERFACIAL ENERGY (σ), GRAIN BOUNDARY ENERGY (σ_{gb}), GIBBS-THOMSON COEFFICIENT (τ) AND ROUGHNESS PARAMETER (α)

Alloy	χ_{PCM}	MP	ΔH (J/mol)	ΔS (J/ mol/K)	α	$\sigma \times 10^2$ (J/m ²)	$\sigma_{gb} \times 10^2$ (J/m ²)	ΔS_V (kJ/ m ³ /K)	ΔH_V	$\tau \times 10^6$ Km
A1	0.1	163	27730	63.600	7.649	36.175	72.252	383.316	167.126	9.439
A2	0.15	160	27695	63.960	7.693	36.685	73.370	392.411	169.914	9.348
A3	0.20	157	27660	64.325	7.736	37.195	74.490	401.874	172.806	9.255
A4	0.25	154	27625	64.695	7.781	37.705	75.410	411.730	175.809	9.157
A5	0.30	150	27590	65.234	7.846	38.215	76.430	422.938	178.903	9.035
E	0.45	140	27485	66.549	8.004	39.745	79.490	457.794	189.069	8.681
A6	0.50	144	27450	65.827	7.917	40.255	80.510	462.194	192.735	8.709
A7	0.55	148	27415	65.118	7.832	40.765	81.530	466.885	196.559	8.731
A8	0.60	152	27380	64.423	7.748	41.275	82.550	471.889	200.553	8.746
A9	0.65	155	27345	63.890	7.684	41.785	83.570	478.334	204.727	8.735
A10	0.70	157	27310	63.511	7.639	42.295	84.590	486.265	209.094	8.697
A11	0.75	162	27275	62.701	7.541	42.805	85.60	491.190	213.668	8.714
A12	0.85	167	27205	61.829	7.436	43.825	87.650	507.947	223.497	8.627
A13	0.87	168	27191	61.657	7.416	44.029	88.058	511.523	225.582	8.607
A14	0.92	171	27156	61.162	7.356	44.539	89.078	520.227	230.981	8.561
PCM		168.7	27100	61.353	7.379	45.356	90.712	543.853	240.220	8.339
PB		174	27800	62.192	7.480	36.175	70.350	361.498	161.842	10.006

TABLE II: PARTIAL AND INTEGRAL MISSING OF GIBBS FREE ENERGY (ΔG^M), ENTHALPY (ΔH^M) AND ENTROPY (ΔS^M) OF PCM-PB SYSTEM

<i>Alloy</i>	ΔG_{PCM}^{-M} J/mol	ΔG_{PB}^{-M} J/mol	ΔG^{-M} J/mol	ΔH_{PCM}^{-M} J/mol	ΔH_{PB}^{-M} J/mol	ΔH^{-M} J/mol	ΔS_{PCM}^{-M} J/mol/k	ΔS_{PB}^{-M} J/mol/k	ΔS^{-M} J/mol/k
A1	-360.677	-680.031	-648.095	7985.663	-297.242	531.048	19.143	0.875	2.701
A2	-548.274	-869.390	-821.226	6281.933	-280.797	703.612	15.772	1.351	3.514
A3	-734.666	-1053.20	-989.493	5019.328	-253.826	800.804	13.380	1.855	4.160
A4	-920.890	-1236.847	-1157.857	4000.937	-213.004	840.481	11.525	2.391	4.674
A5	-1171.101	-1479.175	-1386.752	3063.151	-225.076	761.392	10.009	2.965	5.078
E	-2352.415	-2090.425	-2208.320	954.563	-37.770	408.779	6.638	4.970	5.720
A6	-1536.200	-1846.837	-1691.518	866.734	558.177	712.455	5.762	5.762	5.762
A7	-1289.121	-1599.588	-1428.831	805.044	1197.066	981.453	4.970	6.638	5.720
A8	-1039.187	-1352.251	-1164.412	766.758	1886.862	1214.799	4.247	7.618	5.595
A9	-851.523	-1163.950	-960.872	683.211	2572.717	1344.538	3.581	8.728	5.382
A10	-725.729	-1041.045	-820.323	550.553	3263.993	1364.585	2.965	10.009	5.078
A11	-417.354	-417.354	-417.354	625.670	4278.428	1538.858	2.391	1.525	4.674
A12	-108.647	-428.736	-156.660	486.535	6511.524	1390.283	1.351	5.772	3.514
A13	-46.197	-367.747	-87.998	465.642	7112.959	1329.793	1.157	16.962	3.211
A14	-124.400	-180.879	-128.918	446.661	9139.946	1142.123	0.693	20.998	2.317

TABLE III: PARTIAL AND INTEGRAL EXCESS GIBBS FREE ENERGY (g^E), ENTHALPY (h^E) AND ENTROPY (s^E) OF PCM-PB SYSTEM

<i>Alloy</i>	g_{PCM}^{-E} J/mol	G_{PB}^{-E} J/mol	g^E J/mol	h_{PCM}^{-E} J/mol	h_{PB}^{-E} J/mol	h^E J/mol	s_{PCM}^{-E} J/mol/k	S_{PB}^{-E} J/mol/k	s^E J/mol/k
A1	7985.663	-297.242	531.048	130861.934	-10239.314	3870.810	281.826	-22.802	7.660
A2	6281.933	-280.797	703.612	76692.150	-9461.348	3461.676	162.610	-21.202	6.369
A3	5019.328	-253.826	800.804	49653.452	-8584.420	3063.154	103.800	-19.372	5.262
A4	4000.937	-213.004	840.481	33531.338	-7588.208	2691.678	69.157	-17.272	4.335
A5	3063.151	-225.076	761.392	22477.873	-6548.335	2159.527	45.897	-14.948	3.305
E	954.563	-37.770	408.779	4413.444	-2016.127	877.179	8.735	-4.790	1.134
A6	866.734	558.177	712.455	1814.225	1114.254	1464.239	2.272	-1.333	0.469
A7	805.044	1197.066	981.453	307.595	4946.224	2394.978	1.181	8.905	1.656
A8	766.758	1886.862	1214.799	-2071.317	9742.834	2654.343	-6.678	18.484	3.387
A9	683.211	2572.717	1344.538	-3669.344	15702.045	3110.642	-10.169	30.675	4.126
A10	550.553	3263.993	1364.585	-5139.055	23427.821	3431.007	-13.231	46.892	4.805
A11	625.670	4278.428	1538.858	-6123.745	35114.195	4185.740	-15.515	70.886	6.085
A12	486.535	6511.524	1390.283	-8163.520	79497.669	4985.658	-19.659	165.877	8.171
A13	465.642	7112.959	1329.793	-8514.674	96576.718	5147.206	-20.363	202.865	8.656
A14	446.661	9139.946	1142.123	-9284.871	177059.947	5622.714	-21.917	378.198	10.092

TABLE IV: VALUE OF ΔG_V DURING SOLIDIFICATION FOR PCM-PB SYSTEM OF DIFFERENT UNDERCOOLINGS (ΔT)

Alloy ΔT	$\Delta G_V (J/cm^3)$					
	1.0	1.5	2.0	2.5	3.0	3.5
A1	0.383	0.574	0.766	0.957	1.149	1.340
A2	0.392	0.588	0.784	0.980	1.176	1.372
A3	0.402	0.603	0.804	1.005	1.206	1.407
A4	0.411	0.616	0.822	1.027	1.233	1.438
A5	0.423	0.634	0.846	1.057	1.269	1.480
E	0.457	0.685	0.914	1.142	1.371	1.599
A6	0.462	0.693	0.924	1.155	1.386	1.617
A7	0.467	0.700	0.934	1.167	1.401	1.634
A8	0.472	0.708	0.944	1.180	1.416	1.652
A9	0.478	0.717	0.956	1.195	1.434	1.673
A10	0.486	0.729	0.972	1.215	1.458	1.701
A11	0.491	0.736	0.982	1.227	1.473	1.718
A12	0.508	0.762	1.016	1.270	1.524	1.778
A13	0.511	0.766	1.022	1.277	1.533	1.788
A14	0.520	0.780	1.040	1.300	1.560	1.820
PCM	0.544	0.816	1.088	1.360	1.632	1.904
PB	0.361	0.541	0.722	0.902	1.083	1.263

TABLE V: VALUE OF CRITICAL SIZE OF NUCLEUS (r^*) AT DIFFERENT UNDERCOOLINGS (ΔT)

Alloy ΔT	$r^* (nm)$					
	1.0	1.5	2.0	2.5	3.0	3.5
A1	188.74	125.82	94.37	75.49	62.91	53.92
A2	186.96	124.64	93.48	74.78	62.32	53.41
A3	185.10	123.40	92.55	74.04	61.70	52.88
A4	183.14	122.09	91.57	73.25	61.04	52.32
A5	180.70	120.46	90.35	72.28	60.23	51.62
AE	173.62	115.74	86.81	69.44	57.87	49.60
A6	174.18	116.12	87.09	69.67	58.06	49.76
A7	174.62	116.41	87.31	69.84	58.20	49.89
A8	174.92	116.61	87.46	69.96	58.30	49.97
A9	174.70	116.46	87.35	69.88	58.23	49.91
A10	173.94	115.96	87.97	69.57	57.98	49.69
A11	174.28	116.18	87.14	69.71	58.09	49.79
A12	172.54	115.02	86.27	69.01	57.51	49.29
A13	172.14	114.76	86.07	68.85	57.38	49.18
A14	171.22	114.14	85.61	68.48	57.07	48.92
PCM	166.78	111.18	83.39	66.71	55.59	47.65
PB	200.12	133.41	100.06	80.04	66.70	57.17

TABLE VI: VALUE OF ΔG^* FOR ALLOYS OF PCM-PB SYSTEM AT DIFFERENT UNDERCOOLINGS (ΔT)

Alloy ΔT	$\Delta G^* \times 10^{16} (J)$					
	1.0	1.5	2.0	2.5	3.0	3.5
A1	53.983	23.992	13.495	8.637	5.998	4.406
A2	53.719	23.875	13.429	8.595	5.968	4.385
A3	53.385	23.726	13.346	8.541	5.931	4.357
A4	52.981	23.547	13.245	8.476	5.886	4.324
A5	52.275	23.233	13.068	8.364	5.808	4.267
E	50.194	22.308	12.548	8.031	5.577	4.097
A6	51.163	22.739	12.790	8.186	5.684	4.176
A7	52.070	23.142	13.017	8.331	5.785	4.250
A8	52.909	23.515	13.227	8.465	5.878	4.319
A9	53.425	23.744	13.356	8.548	5.936	4.361
A10	53.612	23.827	13.403	8.578	5.956	4.376
A11	54.466	24.207	13.616	8.714	6.051	4.446
A12	54.661	24.293	13.665	8.745	6.073	4.462
A13	54.655	24.291	13.663	8.744	6.072	4.461
A14	54.699	24.310	13.674	8.751	6.077	4.465
PCM	52.855	23.491	13.213	8.456	5.872	4.314
PB	60.696	26.976	15.174	9.711	6.744	4.954