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Solidification Behavior of Binary Alloys of Nicotinamide-P-Dimethylaminobenzaldehyde Drug System

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Solidification behaviour of Nicotinamide(NA)–p-Dimethylaminobenzaldehyde(MAB) binary drug system have been investigated and reported in this paper. The solid-liquid equilibrium (SLE) data determined by Thaw-melt method with their corresponding compositions construct the solid-liquid phase diagram which suggests simple eutectic of 0.857 mole fraction of MAB at 70 °C is followed by the binary system. The thermodynamic properties such as entropy of fusion, mixing and excess thermodynamic functions of all alloys (A1- A11) have been studied using their enthalpy of fusion values. Gibbs-Duhem equation resolves the quantitative identity of mixing chemical potential, activity and activity coefficient of a component in the binary mix. Interface morphology of the alloys follows the Jackson's surface roughness (α) theory and predicts the faceted growth ($\alpha > 2$) proceeds in all the alloys. The microstructure of pure components of taken binary system having high entropy of fusion as well as the binary organic eutectic of NA-MAB drug system has also been discussed in the present investigation.

Keyword: Phase Diagram, Mixing and Excess Thermodynamic Properties, Roughness Parameter, Microstructure.

1. Introduction

The process of solidification plays a vital role in obtaining homogeneous materials of high quality. The thermal, mechanical, structural and atmospheric stabilities of materials are a subject of potential industrial interest. It has been a great dream of scientists for a long time to find the techniques which control the properties of cast structures because the structure formed immediately after solidification determines the properties of the final products. Due to various difficulties encountered, while working with metallic systems, the interesting idea of observing the solidification phenomena using transparent model is being worked with great enthusiasm for material scientists [1, 2] all over the world. It has been found that at small driving forces, the existence of rough interface leads the necessity for stepwise growth. Only at sufficiently high deriving forces a surface can advance uniformly. Due to low transformation temperature, ease of purification transparency, wider choice of materials and minimized convection effects the organic system are more suitable than those of metallic systems for physicochemical scientific investigation. Formation of molecular complexes of an organic compound with other compounds is due to molecular interactions between them. It is important because the ability to control physical, chemical and pharmaceutical [3, 4] properties without changing the parent covalent bonds. In recent years, research work may extend on the design of pharmaceutical materials [5] by directing molecular association of different components in the solid state to form binary/ternary materials of potential interest.

Nicotinamide, known also as niacin derivative, or vitamin B_3 component, is a water soluble reactive moiety of the coenzyme nicotinamide adenine dinucleotide (NAD) [6]. The physiological function and pharmaceutical action of NAD and NADP is to function as enzymes in a wide variety of enzymatic oxidation-reduction reactions essential for tissue respiration, lipid metabolism

and glycogenolysis. It is anti HIV [7], anti M. tuberculosis [8], anti inflammatory [9] and anti Pellagra [10] agent. Nicam gel [11] is most effective when applied to the skin, which helps to reduce the inflammation and redness of inflammatory acne. P-Dimethylaminobenzaldehyde [12] is a Yellow-white powder and soluble in water. It is used as an Ehrlich's reagent [13] to detect urobilinogen in fresh cool urine. This compound plays an important role in differentiating between serum eruptions and true scarlet fever. Nicotinamide (NA)-p-imethylaminobenzaldehyde (MAB) drug system was selected for the solid-liquid equilibrium phase diagram, mixing and excess thermodynamic functions and investigation of microstructures of eutectic and non-eutectic drug alloys. The excess thermodynamic quantities (g^E, h^E and s^E) have been determined by computing heat of fusion data and activity coefficient of the component in binary mix. These values highlight the ordering, stability and structure of eutectic and non-eutectic alloys. The thermodynamic mixing functions describe about the nature of mixing of the components during alloying. The negative value of integral mixing function, ΔG^M for eutectic and some non-eutectic alloys favours the spontaneous mixing.

2. Experimental Procedure

Nicotinamide (Thomas Baker, Bombey), p-Dimethylaminobenzaldehyde (Thomas Baker, Bombey) were directly taken for investigation. The melting point (experimental value) of nicotinamide, p-dimethylaminobenzaldehyde was found to be 128 °C and 77 °C respectively. The solid-liquid equilibrium data of NA-MAB system was determined by the Thaw-melt method [14, 15]. Mixtures of different composition were made in glass test tubes by repeated heating and followed by chilling in ice. The melting and thaw temperatures were determined in a Toshniwal melting point apparatus using a precision thermometer this could read correctly up to ± 0.1 °C. The heater was regulated to give above 1 °C increase in temperature in every five minutes.To study the microstructure of the pure components and eutectics, a small amount of sample was taken on a well washed and dried glass slide and placed in an oven maintained at a temperature slightly above the melting point of the sample. On complete melting a cover-slip was glided over the melt and allowed to cool. After a few minutes, the supercooled melt was nucleated by the solid of the same composition and care was taken to have unidirectional freezing [16]. After the complete freezing, the slide was placed on the platform of a SES, DMS-01, digital microscope and the different regions of the slide were carried out.

3. Results and Discussion

3.1 Phase diagram (SLE) study

The solid-liquid equilibria (SLE) data of Nicotinamide(NA)–p-imethylaminobenzaldehyde (MAB) system determined by the Thaw melt method is reported in the form of temperature-composition curve (Fig.1). NA-MAB system shows the formation of simple eutectic and formed at 70 °C and 0.857 mole fraction of MAB. The melting point of NA (128 °C) decreases on the addition of second component MAB (77 °C) and further attains minimum and then increases. At the eutectic temperature two phases namely a liquid phase L and two solid phases (S_1 and S_2) are in equilibrium and the system is invariant. In the region indicated by L a homogenous binary liquid solution exists while the two solid phases exists below the horizontal line. In the case, in region located on the left side of the diagram a binary liquid and solid NA exist while in a similar region located on the right side of the diagram a binary liquid and the second component of the system co-exist.

$$L \underset{}{\overset{\text{Cooling}}{\rightleftharpoons}} S_1 + S_2$$

The chemical interaction between two components in a binary system leads to an association of molecules in definite quantities. Physical as well as chemical forces are involved in the formation of eutectic and non-eutectic alloys. Thermochemical studies unfold the nature of mixing as well as nature of interaction between components during binary mix.

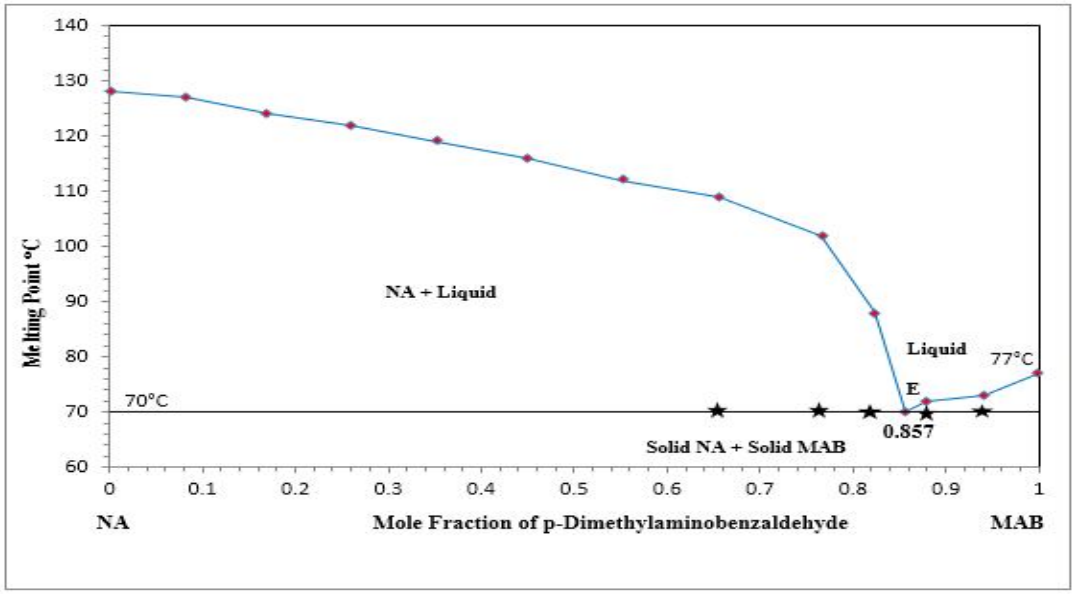

Fig 1: Phase diagram of Nicotinamide-p-Dimethylaminobenzaldehyde system

3.2 Heat of fusion

The values of heats of fusion of eutectic and non-eutectic alloys are calculated by the mixture law using equation [17]

$$(\Delta H)_e = \chi_{NA} \Delta H_{NA} + \chi_{MAB} \Delta H_{MAB} \quad (1)$$

where χ and ΔH are the mole fraction and the heat of fusion of the component indicated by the subscript, respectively. The value of heat of fusion of nicotinamide and 2-methylaminobenzaldehyde is 25400 and 19070 J/mol respectively and all the binary alloys A1-A11, E is reported in Table 1.

Table 1: Phase Composition, Melting Point, Enthalpy of Fusion Values, Entropy of Fusion, Roughness Parameter, Activity and Activity Coefficient

Alloy	χ_{MAB}	MP (°C)	ΔH (J/mol)	ΔS (J/mol/K)	α	a_{NA}	a_{MAB}	γ_{NA}	γ_{MAB}
A1	0.940	73	19449.80	56.21	6.76	0.298	0.927	4.965	0.986
A2	0.880	72	19829.60	57.48	6.91	0.290	0.909	2.420	1.033
E	0.857	70	19975.19	58.24	7.00	0.276	0.875	1.928	1.021
A3	0.823	88	20190.41	55.93	6.73	0.430	1.221	2.429	1.484
A4	0.766	102	20551.22	54.80	6.59	0.590	1.548	2.520	2.021
A5	0.656	109	21247.52	55.62	6.69	0.685	1.731	1.990	2.639
A6	0.551	112	21912.17	56.91	6.85	0.729	1.814	1.623	3.293
A7	0.450	116	22551.50	57.97	6.97	0.791	1.929	1.437	4.287
A8	0.353	119	23165.51	59.10	7.11	0.840	2.018	1.298	5.717
A9	0.260	122	23754.20	60.14	7.23	0.891	2.110	1.204	8.115
A10	0.170	124	24323.90	61.27	7.37	0.926	2.172	1.116	12.779
A11	0.083	127	24874.61	62.19	7.48	0.981	2.269	1.070	27.333

3.3 Activity and activity coefficient

The activity coefficient of components for the systems under investigation has been calculated from the equation [18, 19] given below

$$-ln\chi_i^l\gamma_i^1 = \frac{\Delta H_i}{R}\left(\frac{1}{T_e} - \frac{1}{T_i}\right) \quad (2)$$

where γ_i^1 is activity coefficient of the component *i* in the liquid phase respectively, *Δ*Hi is the heat of fusion of component i at melting point T_i and R is the gas constant. Te is the melting temperature of the binary mix. Values of activity and activity coefficient of all the alloys are given in Table 1.

3.4 Mixing Functions

Integral molar free energy of mixing (*Δ*GM), molar entropy of mixing (*Δ*SM) and molar enthalpy of mixing (*Δ*HM) and partial thermodynamic mixing functions of the binary alloys when two components are mixed together were determined by using the following equations

$$\Delta G^M = RT(\chi_{NA} \ln a_{NA} + \chi_{MAB} \ln a_{MAB}) \quad (3)$$

$$\Delta S^M = -R(\chi_{NA} \ln \chi_{NA} + \chi_{MAB} \ln \chi_{MAB}) \quad (4)$$

$$\Delta H^M = RT(\chi_{NA} \ln \gamma_{NA} + \chi_{MAB} \ln \gamma_{MAB}) \quad (5)$$

$$G_i^{-M} = \mu_i^{-M} = RT \ln a_i \quad (6)$$

where G-iM (μ-iM) is the partial molar free energy of mixing of component i (mixing chemical potential)in binary mix. and γi and ai is the activity coefficient and activity of component respectively. The negative value [20,21] of molar free energy of mixing of alloys (Table 2) suggests that the mixing in all cases is spontaneous. The positive value [22] of ΔG^M infers the mixing in the system is non-spontaneous which may need more energy in the mixing of the components of binary systems. The integral molar enthalpy of mixing value corresponds to the value of excess integral molar free energy of the system favors the regularity in the binary solutions.

Table 2: Value of partial and integral mixing of Gibbs free energy (ΔG^M), enthalpy (ΔH^M) and entropy (ΔS^M) of NA-MAB system

Alloy	ΔG_{NA}^{-M} J/mol	ΔG_{MAB}^{-M} J/mol	ΔG^M J/mol	ΔH_{NA}^{-M} J/mol	ΔH_{MAB}^{-M} J/mol	ΔH^M J/mol	ΔS_{NA}^{-M} J/mol/K	ΔS_{MAB}^{-M} J/mol/K	ΔS^M J/mol/K
A1	-3483.79	-217.94	-413.89	4609.39	-39.95	-239.01	23.39	0.51	1.89
A2	-3547.13	-272.43	-665.39	2534.48	94.24	-387.07	17.63	1.06	3.05
E	-3673.82	-381.40	-852.22	1872.49	58.67	-318.04	16.17	1.28	3.41
A3	-2533.67	599.34	44.80	2663.50	1184.00	-1445.87	14.40	1.62	3.88
A4	-1646.88	1362.14	658.03	2881.44	2193.25	-2354.29	12.08	2.22	4.52
A5	-1203.49	1743.54	729.76	2185.61	3082.51	-2773.97	8.87	3.51	5.35
A6	-1013.47	1907.00	595.71	1549.59	3814.80	-2797.72	6.66	4.96	5.72
A7	-760.10	2124.94	538.17	1173.39	4707.43	-2763.71	4.97	6.64	5.72
A8	-570.07	2288.40	438.97	848.96	5682.05	-2555.04	3.62	8.66	5.40
A9	-380.05	2451.86	356.25	608.79	6875.69	-2238.18	2.50	11.20	4.76
A10	-253.37	2560.83	225.05	361.64	8409.45	-1729.77	1.55	14.73	3.79
A11	-63.34	2724.29	168.03	224.81	11001.42	-1119.27	0.72	20.69	2.38

3.5 Excess Thermodynamic Functions

In order to unfold the nature of the interactions between the components forming the eutectic, non-eutectic alloys and addition compound, the excess thermodynamic functions such as integral excess integral free energy (g^E), excess integral entropy (s^E) and excess integral enthalpy (h^E) were calculated using the following equations The values of $\delta \ln \gamma_1^1 / \delta T$ can be determined by the slope of liquidus curve near the alloys form in the phase diagram. The values of the excess thermodynamic functions are given in Table 3. The value of the excess free energy is a measure of the departure of the system from ideal behavior. The reported excess thermodynamic data substantiate the earlier conclusion of an appreciable interaction between the parent components during the formation of alloys. The positive g^E value for eutectic [23] and non-eutectic

alloys infers stronger interaction between like molecules. The excess entropy is a measure of the change in Configurational energy due to a change in potential energy and indicates an increase in randomness.

$$g^E = RT(\chi_{NA} \ln \gamma_{NA} + \chi_{MAB} \ln \gamma_{MAB}) \quad (7)$$

$$s^E = -R\left((\chi_{NA} \ln \gamma_{NA} + \chi_{MAB} \ln \gamma_{MAB} + \chi_{NA} T \frac{\delta \ln \gamma_{NA}}{\delta T} + \chi_{MAB} T \frac{\delta \ln \gamma_{MAB}}{\delta T} \right) \quad (8)$$

$$h^E = -RT^2 \left(\chi_{NA} \frac{\delta \ln \gamma_{NA}}{\delta T} + \chi_{MAB} \frac{\delta \ln \gamma_{MAB}}{\delta T} \right) \quad (9)$$

and excess chemical potential or excess partial free energy of mixing

$$g_i^{-E} = \mu_i^{-M} = RT \ln \gamma_i^1 \quad (10)$$

Table 3: Value of partial and integral excess Gibbs free energy(g^E), enthalpy(h^E) and entropy(s^E) of NA-MAB system

Alloy	g_{NA}^{-E} J/mol	g_{ACT}^{-E} J/mol	g^E J/mol	h_{NA}^{-E} J/mol	h_{ACT}^{-E} J/mol	h^E J/mol	s_{NA}^{-E} J/mol/K	s_{ACT}^{-E} J/mol/K	s^E J/mol/K
A1	4609.39	-39.95	239.01	306372.94	2107.00	20362.95	872.15	6.21	58.16
A2	2534.48	94.24	387.07	139528.98	3420.31	19753.35	397.09	9.64	56.13
E	1872.49	58.67	318.04	20428.65	-11422.98	-6868.20	54.10	-33.47	-20.95
A3	2663.50	1184.00	1445.87	-8260.06	-15383.77	-14122.87	-30.26	-45.89	-43.13
A4	2881.44	2193.25	2354.29	8075.84	-8843.70	-4884.53	13.85	-29.43	-19.30
A5	2185.61	3082.51	2773.97	62769.49	27165.22	39413.09	158.60	63.04	95.91
A6	1549.59	3814.80	2797.72	56939.15	48026.70	52028.39	143.87	114.84	127.87
A7	1173.39	4707.43	2763.71	43222.70	64802.19	52933.47	108.10	154.49	128.97
A8	848.96	5682.05	2555.04	33837.83	89504.72	53488.24	84.16	213.83	129.93
A9	608.79	6875.69	2238.18	35953.67	155551.98	67049.23	89.48	376.40	164.08
A10	361.64	8409.45	1729.77	37749.94	289250.29	80505.00	94.18	707.41	198.43
A11	224.81	11001.42	1119.27	32625.74	622009.52	81544.59	81.00	1527.52	201.06

3.6 Gibbs-Duhem equation

Further the partial molar quantity, activity and activity coefficient can also be determined by using Gibbs-Duhem equation Using equation (14) a graph between H_{MAB}^{-M} and χ_{MAB}/χ_{NA} (Fig.2) gives the solution of the partial molar heat of mixing of a constituent NA in NA/MAB alloy and plot between ln γ_{MAB} vs χ_{MAB}/χ_{NA} (Fig.3) determines the value of activity coefficient of component NA in binary alloys.

$$\sum \chi_i dz_i^{-M} = 0 \quad (11)$$

$$or \quad \chi_{NA} dH_{NA}^{-M} + \chi_{MAB} dH_{MAB}^{-M} = 0 \quad (12)$$

$$or \quad dH_{NA}^{-M} = \frac{\chi_{MAB}}{\chi_{NA}} dH_{MAB}^{-M} \quad (13)$$

$$or \quad [H_{NA}^{-M}]_{\chi_{NA}=y} = \int_{\chi_{NA}=y}^{\chi_{NA}=1} \frac{\chi_{MAB}}{\chi_{NA}} dH_{MAB}^{-M} \quad (14)$$

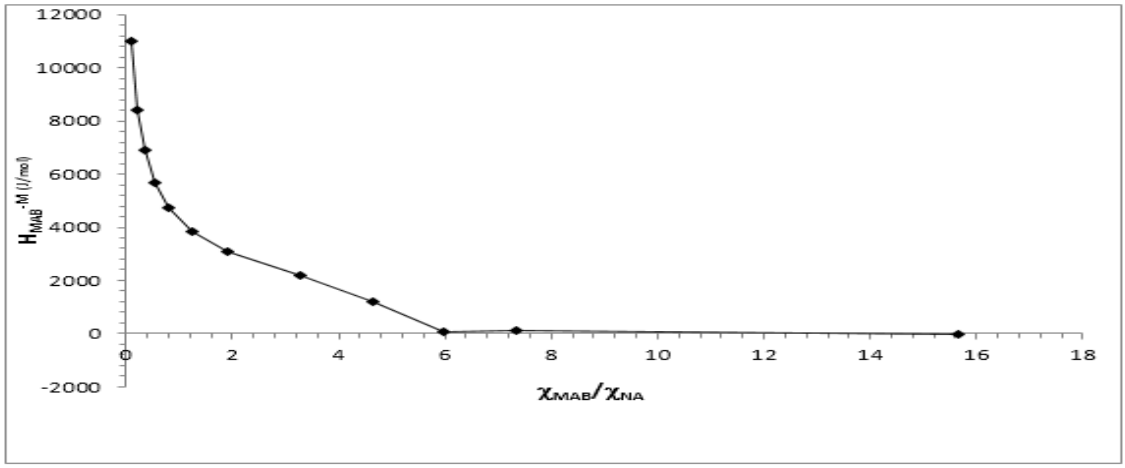

Fig 2: Graphical solution of partial molar enthalpy of mixing of MAB in binary mix

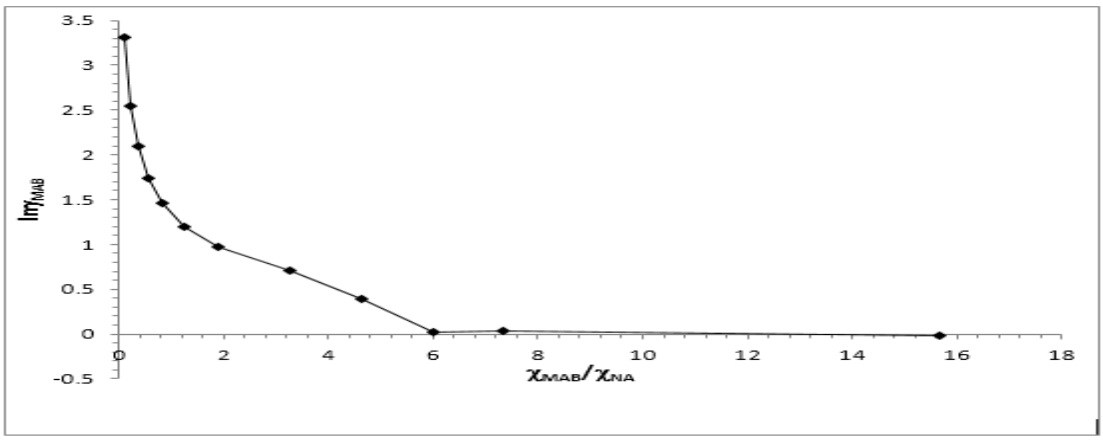

Fig 3: Graphical solution of activity coefficient of MAB in binary mix

4. Interface Morphology

The science of growth has been developed on the foundation of thermodynamics, kinetics, fluid dynamics, crystal structures and interfacial sciences. The solid-liquid interface morphology can be predicted from the value of the entropy of fusion. According to Hunt and Jackson [24], the type of growth from a binary melt depends upon a factor α, defined as:

$$\alpha = \xi \frac{\Delta H}{RT} = \xi \frac{\Delta S}{R} \qquad (15)$$

where ξ is a crystallographic factor depending upon the geometry of the molecules and has a value less than or equal to one. ΔS/R (also known as Jackson's roughness parameter α) is the entropy of fusion (dimensionless) and R is the gas constant. When α is less than two the solid-liquid interface is atomically rough and exhibits non-faceted growth. The value of Jackson's roughness parameter (ΔS/R) of nicotinamide and 2-methylaminobenzaldehyde is 7.62 and 6.55 respectively and all the alloys are given in Table 1. For the entire alloy the α value was found greater than 2 which indicate the faceted [25] growth proceeds in all the cases.

5. Microstructure of Pure Components and Eutectic

The microstructure of pure components NA and MAB and simple eutectic of NA-MAB system are shown in Fig.(4, 5, 6) respectively. The microstructure obtained for the eutectics are of thinly branched, interdendritic, complex regular and irregular type whereas the microstructure obtained for components are of complex and irregular type. A prediction of microstructure of

eutectics can be made from Spengler's equation [26].

$$\theta = \frac{T_1 - T_E}{T_2 - T_E} \qquad (16)$$

where, T_1 and T_2 are the melting temperature of low melting and high melting components,

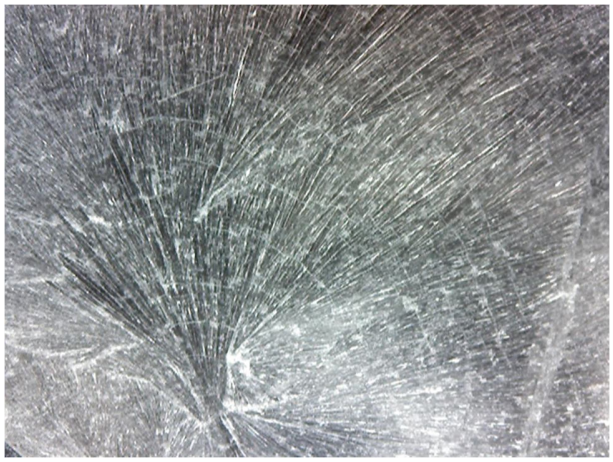

Fig 4: Microstructure of Pure Nicotinamide (NA) x 50

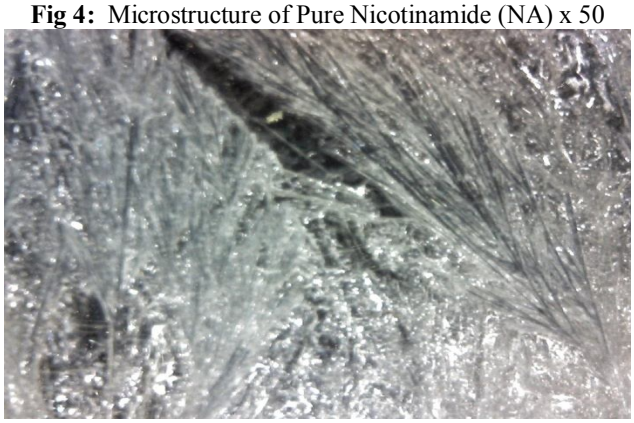

Fig 5: Microstructure of Pure p-Dimethylaminobenzaldehyde (MAB) x 50

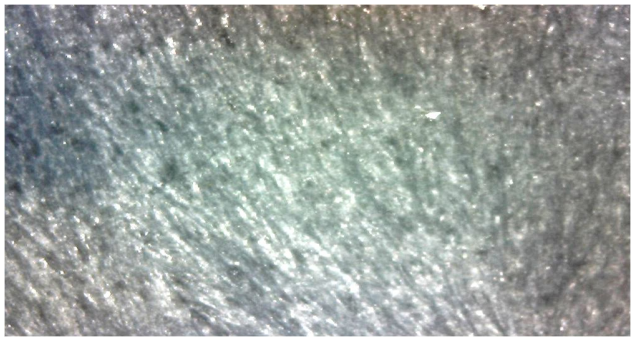

Fig 6: Microstructure of Eutectic of NA-MAB System x 50

respectively; and T_E is the eutectic temperature. The normal eutectics are formed when θ lies between 0.1 and 1.0 but it lies between 0.01 to 0.1 anomalous structure are obtained and when it acquires values less than 0.01, divorced structure are formed. According to Podolinsky *et al.*[27] the surface roughness factor of one component increases or decreases under influence of another component in a binary eutectic, the regular eutectic structure is formed whereas α factor of both eutectic phases decreases under the influence of the components of opposite eutectic phases, anomalous eutectics can be formed. The microstructure of eutectic NA-MAB system having 0.857 mole fraction of second component shows irregular morphology in which the primary arm of a component grows a head of the eutectic interface and interspersed on second phase which is bounded by closed packed plane. The observed microstructure of eutectic are found different from those of pure components.

6. Conclusion

The phase diagram of binary organic alloy of NA-MAB system is found to be simple eutectic. The negative value of molar free energy of mixing of alloys A1, A2 and E suggests that the mixing in all cases is spontaneous and the remaining alloys A4-A11 have the positive value of ΔG^M infers the mixing in the system is non-spontaneous which may need more energy in the mixing of the components of binary systems. The positive g^E value for eutectic and all the non-eutectic alloys infers stronger interaction between like molecules. For all the alloys and eutectic the α value was found greater than 2 which indicate the faceted growth proceeds in all the cases. The computed microstructures the eutectic the system shows irregular morphology in which the primary arm of a component grows a head of the eutectic interface and interspersed on second phase which is bounded by closed packed plane. The observed microstructure of eutectic are found different from those of pure components.

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