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Optimization of stable recipe of semi interpenetrating polymeric network based on the diffusivity and swelling percentage

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Abstract: The hydrophilic nature of semi interpenetrating polymeric network hydrogels (SIPN) bestows a wide spectrum of applications of such systems establishing it as a remarkable material in polymer technology. A SIPN of Polyacrylamide and Glutaraldehyde mediated crosslinked Poly Vinyl Alcohol can be synthesized with variety of combinations of the basic constituents with variable state parameters. In this work we report the fabrication of SIPN matrix hydrogel prepared as small cylinders of length about 2.0 cm and diameter about 0.8 cm in which the constituents are varied in a sequential manner to attain the optimum parametric condition of state of Acrylamide, Glutaraldehyde and Poly Vinyl Alcohol established on the basis of water transport properties based on the diffusivity and swelling percentage studies of the SIPN prepared. We believe this study will pave the way to a new methodology to achieve optimum conditions of precursors in SIPN.

Keywords: Hydrogels, semi-interpenetrating polymeric network, crosslink, swelling percentage, diffusivity.

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1 Introduction: Research over a last few decades has proved that polymeric blends show superior performances over the conventional individual polymers and hence has wide range of applications **[1-4]**. Semi Interpenetrating Polymer Network (SIPN), refers to a specific kind of polymer blend where one polymer is crosslinked in the presence of the other **[5].** Hydrogels made in such a manner have captured much more attention compared to other such blend systems due to their hydrophilic nature offering special ability to hold large amount of water and bio-active compounds without dissolution and hence enhancing drug delivery. Preparation of a SIPN involves a thorough knowledge of the optimum composition of each of the constituents. This study is executed aiming to obtain a stable recipe of the SIPN made from polyvinyl alcohol (PVOH), glutaraldehyde and acrylamide monomer by varying the constituents based on the interrelation of swelling percentage and diffusivity calculated for every individual sample. To delineate the path of achieving the mentioned proposition this paper is oriented in three sections – (1) Synthesis of the material, (2) Characterization of its physical properties and (3) Analysis of the morphological properties.

1. Experimental: First, we report about the synthesized SIPN which is evolved by the polymerization of Acrylamide monomer forming a network with Poly Vinyl Chloride structure with Glutaraldehyde as cross linker and ammonium persulphate as reaction initiator for the acrylamide monomer.

To fabricate the SIPN matrix four solutions were prepared [Solution of polyvinyl alcohol (I), solution of Acrylamide Monomer (II), solution of glutaraldehyde (III), ammonium persulphate solution (IV)] with high performance liquid chromatography (HPLC) water at different specific temperatures commensurating for the system and then brought to room temperature.

A number of steps were followed in a sequential manner with the mixtures of (I), (II), (III) and (IV). Definite proportions of above four solutions were mixed and vortexed for a definite time at definite speed. After the proper dispersion of the mixture the fusion tubes acquired from vortex meter were kept thoroughly immersed in the temperature bath for a definite amount of time at a specific temperature and after bringing those out from the temperature bath cooled to stabilize for a finite time. At the end the fusion tubes were cracked tenderly and the SIPN matrix was retrieved without any damage.

To optimize the stable recipe of semi IPN the recipes of the components were varied and for each of the sample different sets of ensemble experiments were carried out. With the data of these experiments different physical properties like swelling percentage, diffusivity etc. for the SIPN was estimated.

2. Results and discussion:

While preparing semi interpenetrating polymeric network Acrylamide monomer plays the most crucial role other than PV(OH). Therefore with four different varied recipe with different concentrations of Acrylamide monomer i.e., 0.0 %, 5.0 %, 10.0 %, 12.5 % four different SIPN matrix were prepared and the percentile change in mass in water of each of the matrix were calculated.

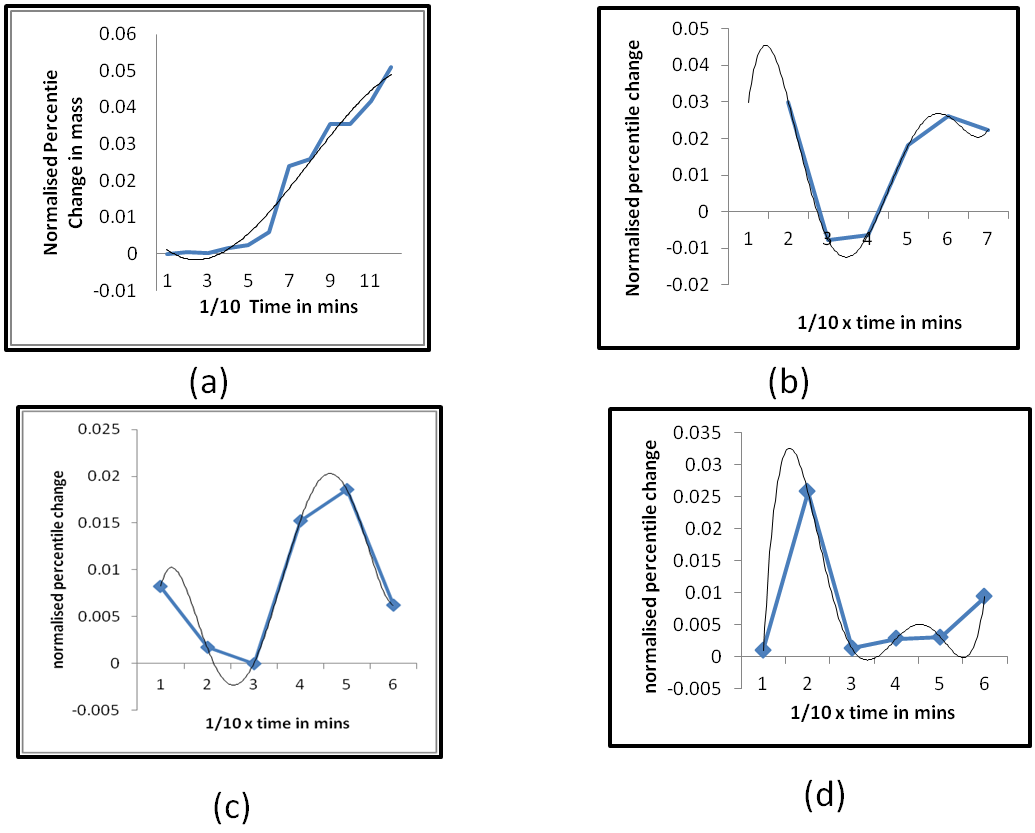

Figure (1): Acrylamide Monomer(AM) conc. (a) 0.0 %, (b) 5.0 %, (c) 10.0 %, (d) 12.5 %.

A set of experiments were performed to determine which recipe of PV(OH) and Acrylamide monomer combination forms the best hydrogel. It was checked by normalized percentile.

Water adsorption study: To calculate the water adsorption the swelling percentage of each of the fabricated matrix has been calculated by the following equation:

$$SwellingPercentage = \frac{(FinalWeight - InitialWeight)}{(InitialWeight)} \times 100$$

From the above four figures we find that the variation trends in figure (b, c, and d) bears some similarity but that in (a) the figure is different. This is probably because of the reason that the acrylamide percentage of 0.0 % in those blends and in absence of acrylamide the polymer matrix is not a semi IPN anymore. Thus we are heading to the exact SIPN matrix structure by combining a finite amount of acrylamide.

As we know due to the hydrophilic nature of the functional group present in the semi IPN generates this unique water adsorption property of this hydrogel whereas the cross linking between network chain prevent them from dissolution **[6]**. Swelling is basically a process of the phase transition in which due to the interaction of solvent molecule with the polymer matrix (During this interaction the functional group of semi IPN forms hydrogen bond and dipole bonds with the solvent molecules) the entropy of the system increases. When the network is swollen by absorption of water, the chain between network junctions are required to assume elongated configuration and a force akin to the elastic attractive force in matrix consequently develops in opposition to the swelling process. As swelling proceeds, this force increases and diluting force decreases. Ultimately a state of equilibrium is reached in which the competing interaction between these two forces is in balance and the entropy attains a constant value. This actually happened due to the Rubber Elasticity **[7],** because the fabricated SIPN, primarily which is a hydrogel, has rubber like structure.

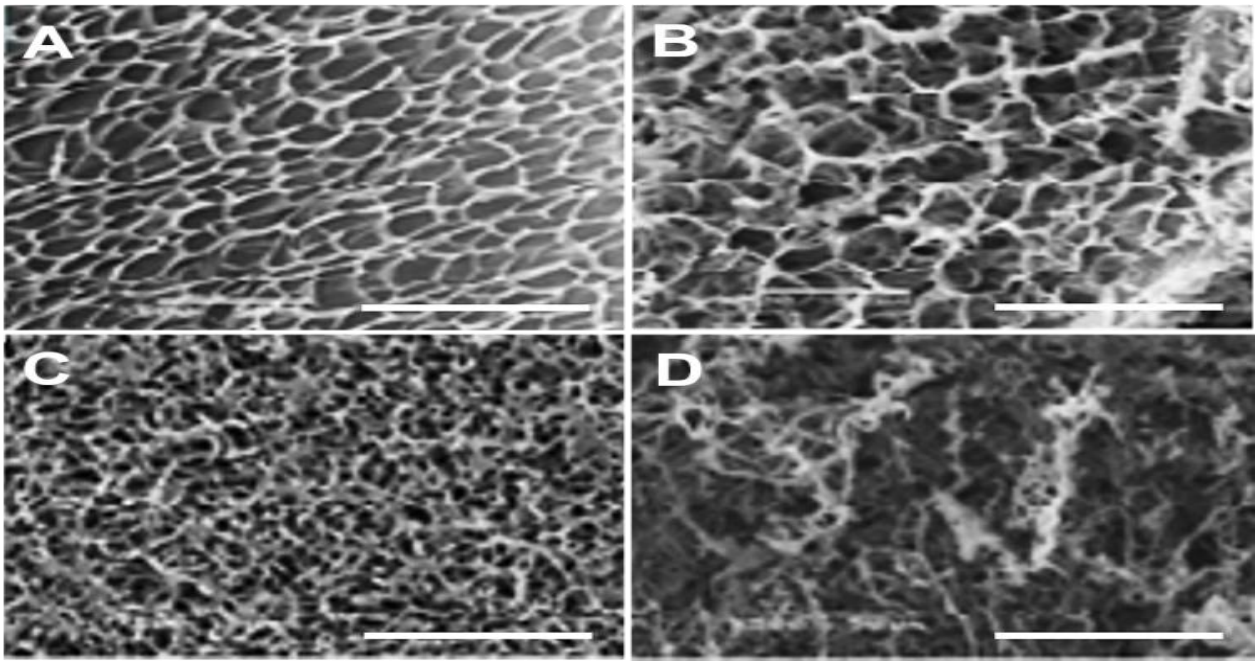

Figure (2): SEM images of (A) Polymeric network with acrylamide monomer (AM) 0.0 %, (B) Polymeric network with AM 5.0 %, (C) Polymeric network with AM 10.0 %; and (C) Polymeric network with AM 12.5 %. Scale bar represent 500 nm.

In our experiment we have measured the normalized percentile change in mass of each of the fabricated semi IPN matrix prepared with different concentrations of acrylamide until it reaches the equilibrium swelling state. While characterizing the swelling properties of the material from above three graphs it is observed that with the increase in concentration of acrylamide there is a consequent variation which was formed following the pattern of a harmonic wave with a phase factor and the most stable one are those with acrylamide 12.5 %.

Figure (2) shows the SEM micrograph image of each of the prepared sample synthesized with different concentration of acrylamide monomer (AM). As it can be seen from the above figure with increase in AM concentration 0.0 % to 10 % the area of the phase domains differentiated by pore boundaries are gradually decreasing which finally obtain an almost homogeneous surface for the AM conc. of 12.5 %. This observation complements well with our experimental result obtained from water absorption studies. This can be conjectured as due to the increase in porosity of interior network structure of the hydrogels with the decrease of the crosslinking level. Therefore, for the optimum concentration of acrylamide as we explored in our study i.e. 12.5 % we get an almost homogeneous surface with comparatively reduced porous structure.

It was observed that the mass, length and diameter of SIPN changes with the changes of concentration of AM. From the values of FWHM (Full Wave at Half Maxima) of the Gaussian distribution of the three different parameters mass, length and diameter of the SIPN matrix fabricated, we conclude that the most stable recipe of the SIPN samples indigenously fabricated in our laboratory are characterized by the mass, length and diameter as 0.27257 gm, 2.006 cm and 0.803 cm respectively.

The variation in mass, length and radius are different for different cooling time in the preparation with exactly same recipe. Actual variation in the morphology is quite apparent from the macroscopic view of the SIPN where it slowly transforms from a rubbery to glassy structure with increase of cooling time. This can be conjectured as - when the polymer is cooled it starts to shrink. The polymer is bulk, not a thin film. The molecules are arranged in different molecular planes. But during the shrinking, molecules in one plane eventually penetrates into the other molecular plane. This result in different molecular configuration. Thus the difference in configuration is the cause of the changes in the morphology.

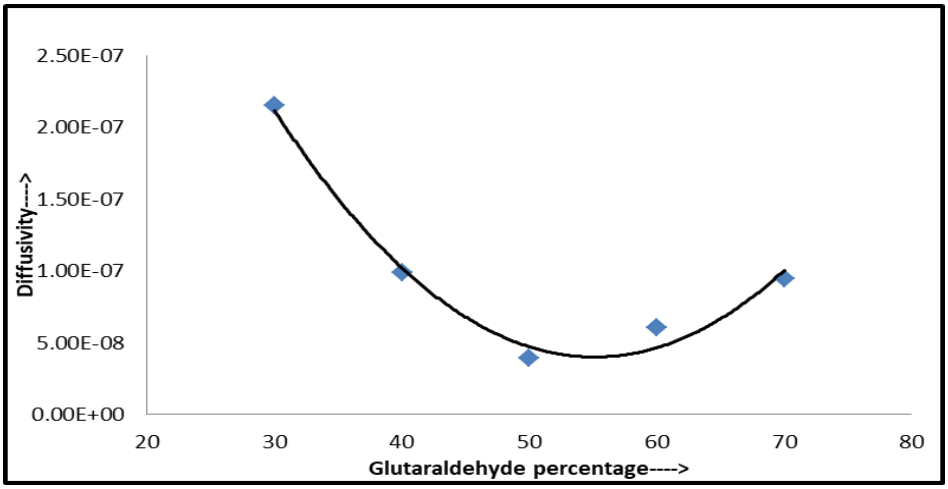

Figure (3): Variation of water diffusivity in SIPN with glutaraldehyde percentage.

Diffusion experiment with polymer at various concentrations of glutaraldehyde was performed from which we have found that the maximum diffusivity can be found at 30 % of glutaraldehyde concentration and minimum value is at 50 % of glutaraldehyde concentration. Thus we can conclude

that, since the minimum is found at 50 % of glutaraldehyde concentration that is the most stable composition of glutaraldehyde in the recipe.

3. Conclusions: From previous literature results we already know that by varying the compositional ratio of SIPN different physical properties of this material such as- hydrophilicity, responsiveness to numerous stimuli, permeability etc. can be controlled and oriented according to the desired fashion **[8]**. Hence, this study not only guides us towards the optimum formulation in preparing a SIPN but the knowledge of each composition also leaves a scope to articulate their physical properties according to the necessary requirement to get the desired outcome **[9, 10]**.

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