

## SEMI-INTERPENETRATING HYDROGEL SYSTEM BASED ON KATIRA GUM (KG) AND POLYVINYL ALCOHOL: SYNTHESIS AND CHARACTERIZATION

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### ABSTRACT

The semi-interpenetrating network (SIPN) based hydrogel was synthesized from katira gum and polyvinyl alcohol in an aqueous medium through cross-linking by glutaraldehyde. The structure and physical properties of hydrogel were assessed by <sup>1</sup>H NMR, FTIR, TGA, XRD, and DSC. The morphological properties and surface charge of the preparations were studied by SEM and zeta potential, respectively. FTIR ascertained the formation of ether linkages in SIPN hydrogel and the alterations in the amorphous structure were revealed by XRD analysis. The thermograms also reflected the formation of effective cross-linkages in synthesized hydrogel as revealed by changes in peak positions of glass-transition temperature. Outcomes of the study disclosed that the SIPN hydrogel of katira gum could be synthesized by applying polyvinyl alcohol and glutaraldehyde. The developed SIPN may be potentially applied in drug delivery, tissue engineering, ion exchange, cell proliferation, dye removal, and heavy metal ions removal from wastewater.

**Keywords:** Katira Gum (KG), Polyvinyl Alcohol, Glutaraldehyde, Semi-Interpenetrating Network, *Cochlospermum religiosum*.

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### INTRODUCTION

The gummy exudates obtained from *Cochlospermum religiosum* (family *Cochlospermaceae*) are heteropolysaccharides in nature. These exudates are also commonly called 'Katira Gum' (KG). It is typically grown in the tropical region of Southeast Asia and in Indian subcontinent. The gummy exudates have wide applications in medical, food, biomedical, pharmaceutical, water treatment, etc.<sup>1,2</sup> It has pronounced physical and chemical properties due to its diverse functionality.<sup>3,4</sup> The natural polysaccharides carry some innate qualities like renewability, non-toxicity, biodegradability, biocompatibility, etc. These properties make them eligible materials for puffiness, suspending mediators, emulsifiers, preservatives, film-forming agents, etc.<sup>5</sup> Chemically, it consists of D-galacturonic acid in a linear chain along with L-rhamnose, and D-galactose with little keto-hexose.<sup>6,7</sup> As a result of remarkable development in biotechnology, its applications in industries have led the way for the utilization of natural polymers. Hence, new materials are being developed in composites, interpenetrating networks (IPNs), semi-IPN (SIPNs), grafted composites, etc. from natural polysaccharides for sustainable development. The possibilities to tune the properties of IPN and SIPN are enormous, and it can be performed using natural, synthetic or semi-synthetic polymers evident from literature reports.<sup>8</sup> SIPN contains one polymer interlaced into another cross-linked polymer without chemical bonds as the network cross-linking is done in the presence of another linear polymer.<sup>9</sup> The generation of SIPNs introduces some entirely new properties in the multifunctional cross-linked polymer, even better than individual polymers.<sup>10</sup> In the past few decades, such materials have been applied in various fields such as metal ion adsorption, dye sequestering, etc.<sup>11,12</sup>

Natural polysaccharides have some limitations such as uncontrolled swelling, syneresis, weak internal structure, the effect of pH, etc. for their use in industries. To overcome such limitations, the natural polysaccharides are being explored in modified forms. Like other natural polymers, KG is also being appreciated in numerous applications due to its prevalent properties.<sup>3,4</sup> Water pollution from harmful

materials like heavy metals, fragment materials, and colour has been a severe environmental issue related to human health. Jana *et al.*<sup>3</sup> prepared KG-based pH-responsive and reusable hydrogels to adsorb organic dyes from water. Ruhidas *et al.*<sup>13</sup> used 1% w/v KG as a matrix-forming drug excipient for a novel drug delivery system and release retarding material in microsphere formulations. Biodegradable hydrogels and grafted polysaccharides have also been used to remove metals and dyes from waste water.<sup>3,14,15</sup> Most of the researchers have studied the removal of a single dye from polluted water, however; there are enormous possibilities that industrial discharge and polluted water contain more than one pollutant.<sup>14,15</sup> Mahmoodi *et al.*<sup>16</sup> studied the dye removal from a multi-component system and successfully removed dyes from the single and ternary system with date stones. In the present study, slightly acidic aqueous dispersion of KG and an aqueous solution of polyvinyl alcohol were used to synthesize SIPN hydrogel. The functionalities of prepared formulations were ascertained through FTIR. TGA and DSC affirmed the thermal stability. The amorphous nature of the prepared material was studied by powder X-ray diffractometry. The surface modifications were examined through SEM photomicrographs. The properties of polyvinyl alcohol as an excellent film-forming material having superb adhesive behavior on solid substrates were utilized for hydrogel formation in the current study.<sup>17,18,19</sup> The blending of two or more polymers can be done to develop the unique properties of the polymeric blend. The mixing of natural and synthetic polymers generates a new class of cost-effective, environmentally and industrially applicable materials.<sup>20</sup>

## EXPERIMENTAL

### Material and Methods

Katira Gum, exudates of the bark of *Cochlospermum religiosum* (family *Cochlospermaceae*), was procured from the local market of Haridwar (Uttarakhand), India. Polyvinyl alcohol (PVA), castor oil, and glutaraldehyde (25% v/v aqueous solution) were purchased from Loba Chemie Pvt Ltd., Mumbai (India). Acetone, Acetic acid, glycine, Span 80, n-Hexane, and other chemicals were obtained from Himedia Laboratories Pvt Ltd., Mumbai (India).

### Synthesis of Semi-IPN Hydrogel

SIPN film was prepared via slight modification in the reported method.<sup>21</sup> A 2% w/v dispersion of KG was prepared in 0.1M acetic acid. A 4% w/v polyvinyl alcohol solution was prepared in hot distilled water at 70°C. KG dispersion (14 ml) was mixed with polyvinyl alcohol (6 ml) in a beaker and stirred on a magnetic stirrer at 200 rpm. After 10 min, a 0.2 ml glutaraldehyde was added drop-wise for cross-linking. The stirring was carried out for 2 h at 30°C. The developed hydrogel was cast into petri-dishes for drying in a hot air oven at 40°C to form the film. The hydrogel film was taken out and washed several times with distilled water and glycine to remove unreacted glutaraldehyde. Finally, the hydrogel was dried in a vacuum dryer at 40°C for 24 h and stored.

### FT-IR Analysis

FT-IR spectra for KG powder and prepared hydrogel were studied by ATR method (Agilent technologies) from 4000–650 cm<sup>-1</sup> to compare the changes in position of functional groups and intensities after semi-interpenetrating network formation.

### XRD Analysis

The diffraction patterns of KG powder and hydrogel were studied by an X-ray diffractometer (XPRT-PRO X-ray generator). It was operated at 40 mA and voltage 45 kV with anode copper K-alpha (1.544 Å) in 2 theta range of 5-80° at 5° per min.

### TGA Analysis

The comparative weight losses at different stages for katira gum and hydrogel film were observed by TGA at a heating rate of 5°C/min with a flow rate of 100 ml/min in a nitrogen atmosphere. The results were recorded in the temperature ranging from 0 to 600°C.

### DSC Analysis

The comparative thermal behavior of KG and the binary blend was assessed by DSC (Perkin Elmer-USA, Model: JADE DSC). The temp was varied from 40 to 300 °C at the rate of 10°/ min during the study. The nitrogen gas at the rate of 20 ml/min was purged during the study.

### SEM Analysis

The surface morphology of KG and hydrogel were investigated by SEM (Model: JSM6100, Jeol) with an image analyser.

### <sup>1</sup>H NMR Study

To assess the crosslinking and formation of SIPN; <sup>1</sup>H NMR spectra were also recorded (Bruker 500 MHz, AVANCE III). Approximately, a 500 mg of sample was dissolved in dimethyl sulphoxide at 70°C to record the NMR spectrum. The <sup>1</sup>H-NMR data was processed by Mestrenova<sup>®</sup> software.

### Zeta Potential Analysis

The zeta potential of gum powder and hydrogel was measured by dispersing in demineralized water and taken in clear disposable zeta cell at 25°C (Malvern, Zetasizer Version 7.11). It was operated at count rates 267.1 and 147.0 kcps with a measurement position at 2.00 mm for 10 zeta runs. The measurements were performed three times for each sample and the average values are reported.

## RESULTS AND DISCUSSION

### Crosslinking of KG and Polyvinyl Alcohol with Glutaraldehyde during SIPN Formation

Components of KG have a cyclic furanose structure. A schematic presentation of the chemical structure of KG has been shown in Fig.-1. In this process, glutaraldehyde was added to cross-link natural polymer (KG) with polyvinyl alcohol. During the synthesis process, glutaraldehyde reacted with –OH groups of KG and polyvinyl alcohol. The cross-linking reaction sites in KG might be D-galactose units to form 5 membered rings having acetal linkage since it has *cis* symmetry of both adjacent –OH groups. In contrast, polyvinyl alcohol might form 6 membered ring having acetal linkage after cross-linking. A schematic representation of cross-linking has been shown in Fig.-2. The mechanism of the cross-linking reaction might be acid-catalyzed nucleophilic addition as shown in Fig.-3.

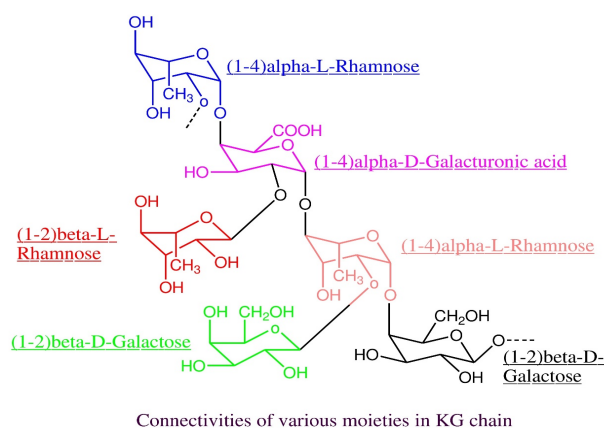


Fig.-1: Chemical Structure of KG

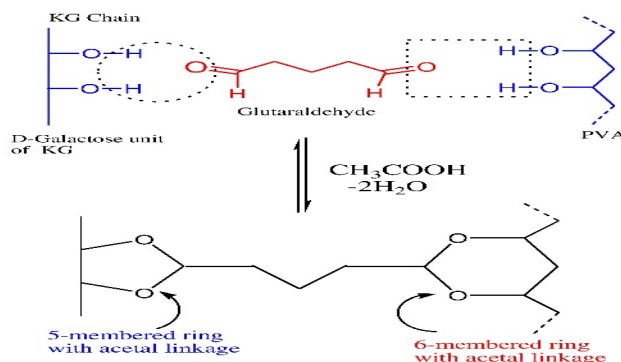


Fig.-2: Crosslinking Reaction of Glutaraldehyde with KG and Polyvinyl Alcohol

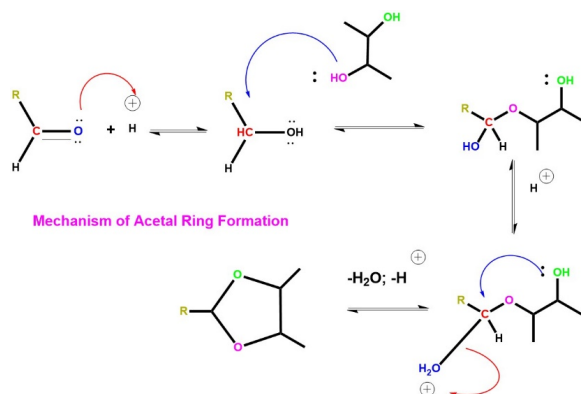


Fig.-3: Nucleophilic Addition of Glutaraldehyde in the Crosslinking Process

The reaction of glutaraldehyde creates ether linkages in structure and results in formation of semi-interpenetrating network. During this process, some of the unreacted glutaraldehyde might have adhered to the surface of the synthetic hydrogel. The traces of glutaraldehyde were washed out by successive washing with distilled water and glycine. Glycine reacts with glutaraldehyde and forms complex. The colour change of Fehling solution assessed the presence of glutaraldehyde as the indicator turns into a red colour after reacting with an aldehyde.

### Characterization of SIPN Hydrogel of KG and Polyvinyl Alcohol

#### Fourier Transform Infrared Analysis

The acquired FT-IR spectra indicated the presence of specific functional groups in samples under observation by absorption band at particular wavenumbers according to vibrational frequencies. Inter and intra-molecular hydrogen bonding were also found in FTIR spectra. The FTIR spectra of semi-IPN hydrogel and KG have been shown in Fig.-4. The addition of glutaraldehyde as cross-linking agent revealed broadband around  $3353\text{ cm}^{-1}$ <sup>18, 21</sup>. The stretching frequency of O-H present in polyvinyl alcohol and glutaraldehyde appeared at  $3550\text{--}3200\text{ cm}^{-1}$  due to inter- and intra-molecular hydrogen bonding. The C-O-C linkage formed during the cross-linking was found at around  $1150\text{ to }1085\text{ cm}^{-1}$ . These significant changes were due to acetal group formation between -OH of KG and polyvinyl alcohol and -CHO of glutaraldehyde.<sup>22</sup> FTIR spectra revealed peaks near  $893$  and  $1156\text{ cm}^{-1}$ , indicating the saccharide structure. Stretching vibrations of C=O from -COOH groups present in KG were observed at  $1745\text{ cm}^{-1}$ .<sup>23, 24, 25</sup> The bands near  $3435\text{ cm}^{-1}$  and  $2924\text{ cm}^{-1}$  were speculated due to stretching of -OH and -CH<sub>2</sub> groups respectively. The bands at  $1607\text{ cm}^{-1}$  and  $1421\text{ cm}^{-1}$  were attributed due to -C=O moiety while the band at  $1037\text{ cm}^{-1}$  could be assigned due to C-O stretching. A peak at  $858\text{ cm}^{-1}$  showed the presence of glycosidic bonding among the monosaccharide units.<sup>26</sup> The bands appearing near  $1039\text{ cm}^{-1}$  might be due to stretching vibrations of -C-OH bonds present in the glucopyranose ring of the KG unit. The sharp peaks at  $1380\text{ cm}^{-1}$  and  $1421\text{ cm}^{-1}$  showed -CH<sub>3</sub> symmetrical deformation mode.<sup>27</sup> The shifting and development of new bands supported the formation of a semi-IPN hydrogel.<sup>28</sup>

#### Morphological Characterization by SEM Analysis

SEM images provide a closer look at the surface morphology of any substance under observation. The SEM images of KG powder and hydrogel are shown in Fig.-5. A significant difference was observed in morphologies of prepared hydrogel and pure KG powder. The SEM image of KG showed a distinctive sliver structure which might be due to intra-molecular H-bonding. Numerous fine particles were embedded firmly in the hydrogel surface. These particles might be responsible for increment in surface area and might be the binding sites for other molecules/ions. Such morphologies of the SIPN blend could make them suitable members for cell adhesion, proliferation and adsorption of metal ions, dyes, etc.<sup>18</sup>

#### XRD Analysis

A powder-XRD investigation was carried out to find out possible alterations in the microstructure of KG-SIPN. The X-ray diffraction spectra of KG and KG-SIPN are shown in Fig.-6. The XRD pattern of KG was similar to amorphous material without any sharp peak while the diffractogram of KG-Hydrogel showed characteristics diffraction peaks of polyvinyl alcohol at  $2\theta=19.6964^\circ$ .

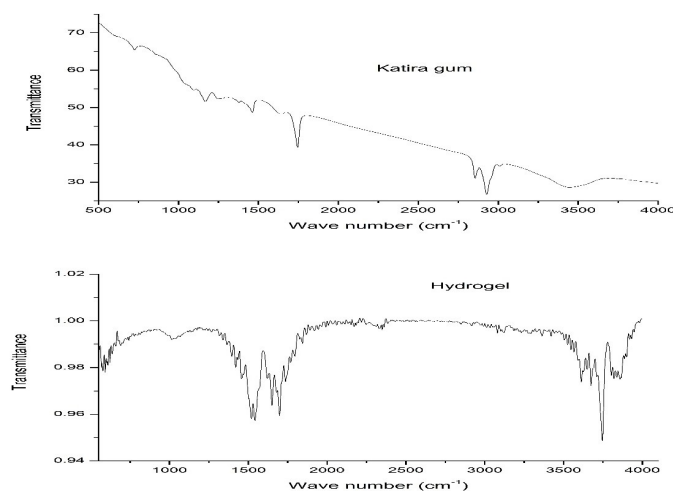


Fig.-4: FTIR Spectrograms of KG and Semi-IPN Hydrogel

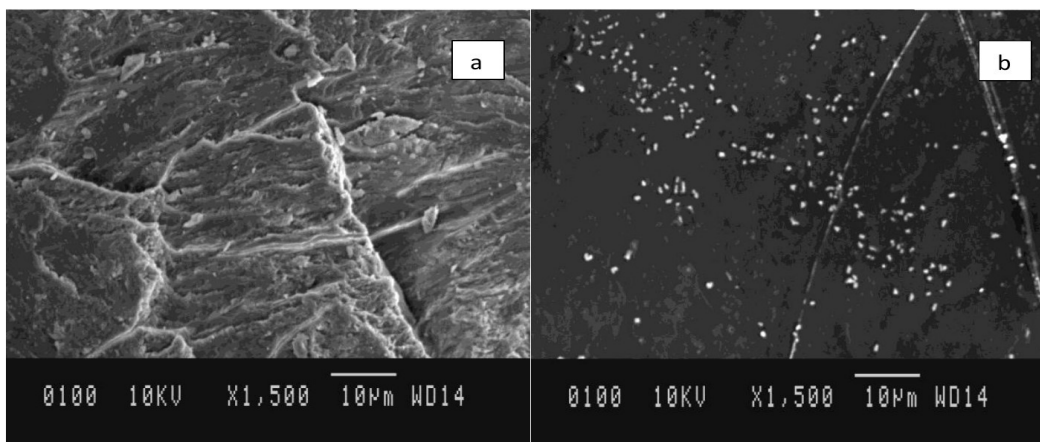


Fig.-5: Surface Morphology of (a) KG and (b) Semi-IPN Hydrogel

Tang *et al.*<sup>29</sup> reported that at around  $2\theta = 19.92^\circ$ ; a strong crystalline peak is observed for PVA with a shoulder peak at  $2\theta = 22.74^\circ$ . Hence, the appearance of characteristic sharp peaks with higher intensity indicated more crystallinity of KG after cross-linking. The strong hydrogen bonding interaction between KG and PVA chains has been reported elsewhere. It may be responsible for intense cross-linking with glutaraldehyde and resulting in a higher degree of cross-linking.<sup>30</sup> The presence of hydrophilic groups in the polymeric structure of KG and polyvinyl alcohol should also result in good miscibility in the binary blend. This also implies that there is an increment in crystallinity after cross-linking. Here, the peak intensity suggests that the amorphous nature of KG was modified after the formation of a semi-IPN hydrogel.<sup>31</sup> The intermolecular hydrogen bonding between KG chains resulting in the cross-linking process is responsible for its semi crystallinity.<sup>32</sup> The crystallinity was increased after cross-linking in the semi-IPN hydrogel. It reflects that the cross-linking occurred at the time of SIPN-based hydrogel formation and KG and PVA were interspersed uniformly.<sup>33</sup>

### TGA Analysis

TGA thermograms of KG-SIPN hydrogel and KG are shown in Fig.-7. In thermogram of KG, an initial 17.55% w/w weight loss was observed from 34.87 to 79.22°C owing to the loss of bonded and unbounded water content. In the second stage, the weight loss was found at 43.47% w/w and it occurred from 238.49 to 278.19°C. The second stage decomposition might be attributed due to the breakage of C-O-C bonds and the elimination of water molecules from the saccharide ring.<sup>34</sup> Final degradation showed 9.26% w/w weight loss at around 383.49°C and the weight loss was attributed due to the formation of carbonaceous material.<sup>35</sup> In semi-IPN hydrogel; a 12.23% weight loss was

observed in the temperature ranging from 31.53 to 93.48°C and a 35.55% w/w weight loss was seen in the second degradation stage that occurred from 251.53 to 281.07°C

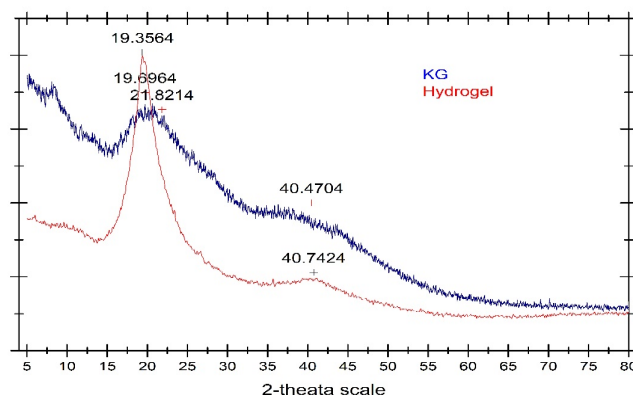


Fig.-6: XRD Diffraction Patterns of Pure KG and Semi-IPN Hydrogel

The final degradation stage showed 30.43% w/w weight loss during the thermal range from 337.19 to 368.70°C which could be due to the breaking of crosslinking. The sharp weight loss in the semi-IPN hydrogel in the final stage would be due to the breaking of the  $-\text{COOH}$  groups of the KG chain and degradation of the SIPN structure.<sup>36</sup> Slower and altered degradation pattern of hydrogel from KG indicated the crosslinking of KG and polyvinyl alcohol with glutaraldehyde to form semi-IPN hydrogel film. Thakur and Arotiba<sup>5</sup> reported a similar thermal degradation pattern of grafted sodium alginate hydrogel system. The degradation of sodium alginate hydrogel indicated a slower weight loss rate in comparison to native sodium alginate and it was considered due to the grafting process.

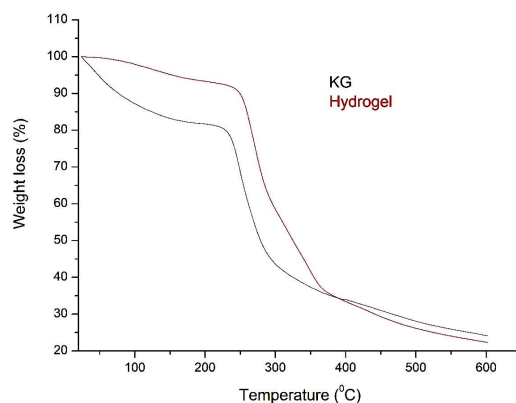


Fig.-7: TGA Thermograms of Pure KG and Semi-IPN Hydrogel

### DSC Analysis

DSC investigation was carried out to observe the miscibility of polymeric materials, thermal firmness, chemical cross-linking process and the changes in glass-transition temperature (tg). DSC thermograms of pure KG and the developed semi-IPN hydrogel are shown in Fig.-8. The thermogram of pure KG showed an endothermic peak at 109.53°C. At this temperature, the peak height and the peak area were found as 29.86 mW and 6015.95 mJ respectively. The enthalpy changes as  $\Delta H$  and  $\Delta H_f$  were found as 1203.19 J/g and 1.20 KJ/mol respectively. However, semi-IPN hydrogel showed an endothermic peak at 115.09°C and the peak height and the peak area were found as 15.97 mW and 2539.38 mJ respectively. The values of  $\Delta H$  and  $\Delta H_f$  for SIPN were found as 507.87 J/g and 0.50 KJ/mol respectively. The need of more energy for the decomposition of semi-IPN indicated the improved thermal firmness of hydrogel. Remarkable miscibility of the polymeric blend was also inferred by the observed thermogram. The shifting of the endothermic peak in the direction of higher temperature due to the entanglement of polymer chains indicated the formation of a semi-IPN structure.<sup>37</sup>

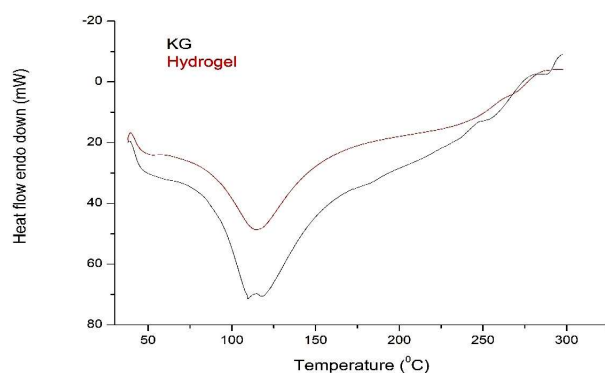
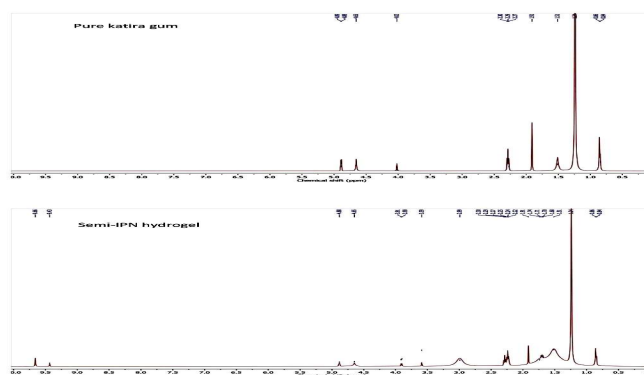


Fig.-8: DSC Thermograms of Pure KG and Semi-IPN Hydrogel

### **<sup>1</sup>H NMR Analysis**

<sup>1</sup>H NMR spectra of native KG showed the deshielded chemical shifts at 4.8, 4.6 and 4.02 ppm due to the protons of –OH groups in different chemical environment. These chemical shifts were also observed in <sup>1</sup>H spectra of semi-IPN hydrogel except 4.02 ppm peak and these might be due to the consumption of protons during cross-linking of KG and PVA chains by glutaraldehyde. Additionally, two highly deshielded singlets at 9.6 and 9.4 ppm were also observed in hydrogel spectra which might be due to protons of –CHO groups connecting KG and PVA chains. The triplet of KG spectra with an intensity ratio of 1:2:1 was split into multiplet with a random intensity pattern. A singlet at 1.51 ppm was also split into a triplet with a 1:1:1 intensity pattern. It was speculated due to the changes in the chemical environment after crosslinking. Apart from these, some new peaks were also observed in SIPN hydrogel spectra which were not present in the spectra of native KG. The appearance of new peaks and improved intensity of existing peaks indicated the modifications that occurred during crosslinking of KG and PVA for semi-IPN formation. The peak pattern of <sup>1</sup>H NMR of native gum katira and hydrogel has been given in Table-1. <sup>1</sup>H NMR spectra of native KG and hydrogel are shown in Fig.-9.

Fig.-9: <sup>1</sup>H NMR Spectra of Pure Gum and Semi-IPN Hydrogel

### **Zeta Potential Analysis**

The apparent zeta potential distribution curve of KG and semi-IPN hydrogel are shown in Fig.-10. The surface charges of an aqueous dispersion of KG were found to be –6.06 mV with the conductivity of 0.264 m S/cm. However, after crosslinking, the surface charge on hydrogel was found to be 0.318 mV with of the conductivity 0.0840 m S/cm. The negative charge would be due to the ionization of –COOH groups in water present in the aqueous dispersion of KG and hydrogel. In curves, two peaks were observed at 59.3 mV with 70.3% area and -142 mV with 29.7% area in the zeta potential of the hydrogel. While in the zeta potential curve of KG, two very closely located peaks were observed at –16.2 mV with 62.0% area and at 10.8 mV with 38% area.

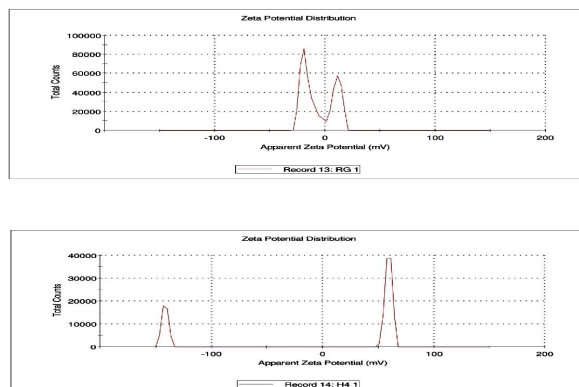


Fig.-10: Zeta Potential of Pure Gum Katira and Semi-IPN Hydrogel

Table-1: Peaks Position of  $^1\text{H}$  NMR in Semi-IPN Hydrogel and Native KG

| Splitting Pattern                                            | $^1\text{H}$ NMR of native KG | $^1\text{H}$ NMR of semi-IPN hydrogel |
|--------------------------------------------------------------|-------------------------------|---------------------------------------|
| Singlet                                                      | -                             | 9.6 ppm                               |
| Singlet                                                      | -                             | 9.4 ppm                               |
| Singlet                                                      | 4.88 ppm                      | 4.88 ppm                              |
| Singlet                                                      | 4.60 ppm                      | 4.6 ppm                               |
| Singlet                                                      | 4.06 ppm                      | -                                     |
| doublet, 1:1 intensity                                       | -                             | 3.9 ppm                               |
|                                                              | -                             | 3.8 ppm                               |
| Singlet                                                      | -                             | 3.59 ppm                              |
| Singlet                                                      | -                             | 2.99 ppm                              |
| Triplet, 1:2:1<br>Multiplet with random<br>intensity pattern | 2.27 ppm                      | 2.30 ppm                              |
|                                                              | 2.29 ppm                      | 2.28 ppm                              |
|                                                              | 2.30 ppm                      | 2.27 ppm                              |
|                                                              |                               | 2.25 ppm                              |
|                                                              |                               | 2.23 ppm                              |
| Singlet                                                      | 1.91 ppm                      | 1.91 ppm                              |
| Singlet                                                      | 1.51 ppm                      | 1.24 ppm                              |
| Singlet                                                      | 1.23 ppm                      | -                                     |
| Doublet, 2:1 intensity                                       | 0.86 ppm                      | 0.86 ppm                              |
|                                                              | 0.84 ppm                      | 0.84 ppm                              |

## CONCLUSION

In the study, aide to formulate KG-SIPN hydrogel was formulated successfully through a cross-linking mechanism using glutaraldehyde as a crosslinker. The cross-linking of KG and polyvinyl alcohol with glutaraldehyde successfully developed semi-IPN hydrogel with improved thermal and surface properties. The hydrogel was found insoluble in aqueous and most organic solvents. FTIR and  $^1\text{H}$  NMR spectrum showed the chemical interaction between KG and PVA. The DSC and TGA analysis revealed more thermal stability of semi-IPN hydrogel. XRD patterns reflected the overall decrement in the amorphous nature of SIPN hydrogel and it would be possible due to cross-linking and modified polymeric chains organization and mechanical properties. SEM images of hydrogel reflected the apparent roughness with increased surface area. The improved surface properties may be responsible for enhancing SIPN utility as adsorbents for dye or heavy metal ion removal from wastewater. The effective cross-linking of KG and polyvinyl alcohol with glutaraldehyde reflected the efficacy of the adopted process in developing semi-IPN hydrogel and it would be a remedy for versatile applications. Hydrogels are hydrophilic with regulated high swelling capacity. Water retention in hydrogel may be a dominant parameter and prerequisite quality for the various environmental applications including wastewater treatment. The functional groups like  $-\text{COOH}$ ,  $-\text{OH}$ , etc. in prepared SIPN hydrogel may be suitable Lewis bases and can serve as chelating agents for binding heavy metal ions from wastewater. The hydrogels also have the

potential for different industrial applications e.g. packaging, tissue engineering, heavy metal ion removal from wastewater and in developing drug delivery devices. Over the recent years, hydrogels have emerged as a hope for such applications due to their biodegradability, biocompatibility and similarity with natural outer cell matrix. In the hydrogel system, the porosity and, consequently, water retention capacity can be tuned successfully. IPNs and SIPNs derived from synthetic and natural polymers may result in bio-compatible and eco-friendly materials.

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