

Molecular Structure

Elsevier Editorial System(tm) for Journal of

Manuscript Draft

Manuscript Number: MOLSTRUC-D-20-04374R1

Title: Polyvinyl alcohol (PVA) mixed green-clay and aloe vera based polymeric membrane optimization: Peel-off mask formulation for skin care cosmeceuticals in green nanotechnology

Article Type: VSI:Nanoarchitectonics

Keywords: Aloe vera; MMT; PVA; Polymeric membrane; Skin mask formulation

Corresponding Author: Dr. Nidhi Asthana,

Corresponding Author's Institution: University of Allahabad

First Author: Nidhi Asthana

Order of Authors: Nidhi Asthana; Kaushik Pal, Ph.D.;D.Sc.(Malaysia);Marie-Curie(Europe);CAS Flw; Kamlesh Pandey, Ph.D.; Alaa A Aljabali, Ph.D.; Murtaza M Tambuwala, Ph.D.

<p align="center">Journal of Molecular Structure – Elsevier (Special Issue – Nanoarchitectonics: From molecules to Advanced nanomaterials) Response to Reviewers Polyvinyl alcohol (PVA) mixed green-clay and aloe vera based polymeric membrane peel-off mask formulation skin care avenue of green nanotechnology First Submitted Manuscript ID: MOLSTRUC-D-20-04374 (The revised version was written in highlighted text in the manuscript)</p>		
<p align="center">Comments from Reviewer # 1</p>		
No.	Comments:	Author Response
1	Title needs a revision. It doesn't reflect clear meaning of the work.	'Title' is amended as suggested mentioned below; "Polyvinyl alcohol (PVA) mixed green-clay and aloe vera based polymeric membrane optimization: Peel-off mask formulation for skin care cosmeceuticals in green nanotechnology"
2	Under section 2, first sentence "we used" should be replaced by "we used the following".	Revised as informed.
3	In page 6, authors claims that the physical stability of the gel solution is 30-40 days. How they confirmed it? Elaborate.	Aloe vera chemical assisted solution to pure gel formation is physically stable up to 30-40 days, only it is possible due to chemical aging. Without proper gel formation uable to get any spectroscopic responses like: XRD, FT-IR etc.
4	Authors should represent fig. 3(a), 3(b) and 3(c) clearly.	We amended Figures 3(a) to (c)
5	Show the XRD pattern in a separate figure, and use the full form of CPS.	We replace XRD patterns separately and CPS is 'Count Per Second'
6	Give the proper representation of a, b, c in XRD pattern. Explanation for compound a,b is missing. Include the physical meaning of (101) and (111) planes shown.	Proper crystallization of (hkl) peak patterns shown as (101) and (111) as mentioned in corresponding (a) and (b) different concentration of Polyvilyle alcohol (PVA) and aloe vera (AV).
<p align="center">Comments from Reviewer # 2</p>		
The manuscript deserves to be published after the minor revision.		
No.	Comments:	Author Response
1	In 'Abstract' language should be improved and abbreviations should be avoid.	Abstract is amended as suggested
2	In the 'Introduction' section cited references are inadequate; more application based green nanotechnology articles should be cited.	We cited current references in the 'Introduction' section.

3	Figures should not be reused from other publications. In case, figures are reused, there will be a copy right infringement. Hence authors are requested to focus on this matter.	All figures are author's own creation of intellectual properties, so not required further copyright permissions from any concern journals.
4	The role of PVA in mixed green-clay and aloe vera based polymeric membrane, should be explain clearly	Film-forming performance was influenced only by the Poly-vinayl alcohol (PVA) concentration, achieving maximum levels at concentrations, as well as the lowest drying time possible and excellent homogeneity of the green clay particles and aloe vera before and after drying.
5	FT-IR spectra (Fig.5) should be detailed more.	The FT-IR is to investigate number and types of bioactive compounds present in Aloe Vera buds grown in North East India at Allahabad region is taken, nondestructive, cost effective and user friendly. Aloe vera was cultivated under normal atmospheric conditions. FT-IR suggested that the presence of acetyl groups is necessary for biological activation, possibly because they cover a number of hydrophilic hydroxyl groups and thus make the molecule more able to cross hydrophobic barriers in the cell. Our significant effort to investigate functional groups present in <i>Aloe vera</i> buds by using FT-IR spectroscopic technique which is simple, cost effective and user friendly technique to investigate functional groups of compounds in sample.
6	'Conclusions' section is correctly written, but a language proof reading is required for better language.	Revised article 'Conclusion' section amended with new finding of aloe vera based research application

Comments from Reviewer # 2

Details presented in depth review of the article entitled 'Polyvinyl alcohol (PVA) mixed green-clay and aloe vera based polymeric membrane peel-off mask formulation skincare avenue of green nanotechnology' are skillfully presented by the author(s). The review article covered all the scientific aspects of the composite membrane as well as its health care applications. The article provides the novelty of relevant research innovations as well as

	<p>profitable statistical tools that aid the conviction of the nature of various compounds based on their spectroscopic responses. Before publishing, the author(s) should pin point and revise the following doubts and questionnaires to help further the readers in easy understanding of the research. Overall the presentation and details of the review report stands and the author should give a chance for further 'Minor Revision' as mentioned in the above points, which are mandatory as per the publication standard of the 'Journal of Molecular Structure'.</p>	
1	'Abstract' is perfectly well written, however some abbreviations leaves the readers in confusion, please make a list of all the abbreviations after the keywords.	We mentioned list of of abbreviation further clear understanding before started 'Introduction' section.
2	'Introduction' page no. 3 'acetylated' should be 'acetylated'.	We amended the typos
3	'Introduction' page no. 3 'properties of antiseptic and analgesic. it contains out of 22 amino acids' should be 'properties of antiseptic and analgesic. It contains out of 22 naturally occurring amino acids'	Revised article all typos we corrected
4	'Introduction' page no. 3 '8 essential amino acids it has 8. Which are antibacterial in nature.' should be '8 essential amino acids it has 8, which are antibacterial in nature.'	Corrected as suggested
5	What's the special reason for choosing the aloe vera by the author?	An emerging herbal product 'Aloe vera' is most widely utilized for skin treatments due to its gel-components and cost-effective and easily available.
6	'Introduction' section should cite with current references based on green chemistry approach of novel nanomaterials applications, as references are not enough cited.	Related aloe vera based application article cited with current references, as suggested by the reviewers.
7	All Figures are extremely informative as well as eye-catching too, and are helpful to the audience to further understand the main theme of the work presented by the author. If those figures are reused, the authors should mention the copyright permissions and are mandatory for the acceptance of the manuscript.	We drawn all figures which are author's own creation of intellectual properties, so not required further copyright permissions from any concern journals.
8	What's the nanoscale dimension of taken SEM image in Figure 2(c)?	The scale under 1 mm in laboratory microscopic scale dimension.
9	'Conclusions' covered all the aspects for detail understanding, however a proof reading grammatical errors, sentence framing and language polishing required by the	Overall manuscript and 'conclusion' section proof reading done thoroughly checked and polishing in revised manuscript accordingly suggested.

	author.	
--	---------	--

**Polyvinyl alcohol (PVA) mixed green–clay and aloe vera based polymeric
membrane optimization: Peel-off mask formulation for skin care
cosmeceuticals in green nanotechnology**

Nidhi Asthana^{1,*}, Kaushik Pal^{2,3,}, Kamlesh Pandey¹, Alaa AA Aljabali⁴, Murtaza. M. Tambuwala⁵, Fernando Gomes de Souza²**

^{1,}National Centre of Experimental Mineralogy and Petrology, University of Allahabad, Allahabad – 211002, India*

^{2,}Laboratório de Biopolímeros e-Sensores/LaBioS Centro de Tecnologia – Cidade Universitária, Federal University of Rio de Janeiro, 21941-901, Brazil.*

^{3,}Wuhan University, 8 East Lake South Road, Wuchang 430072, Hubei Province, China.*

^{4,}Department of Pharmaceutical Sciences, Yarmouk University—Faculty of Pharmacy, Irbid 566, Jordan.*

^{5,}Department of Pharmacy and Pharmaceutical Sciences, University of Ulster, United Kingdom.*

*Corresponding authors:

***Dr Nidhi Asthana** (asthananidhi27@gmail.com)

And,

****Prof (Dr.) Kaushik Pal**, Ph.D.; D.Sc. (Malaysia); Marie-Curie (Europe); Scientist (South Korea); CAS Fellow (China)



Abstract.

An emerging herbal product 'Aloe vera' is most widely utilized for skin treatments due to its gel-components of the plant are known to heal the skin from a variety of minor ailments. In depth experimental attempts with Aloe vera (AV), MMT (Clay) and PVA (Poloy vinyle alcohol) blend gel were prepared using the solution cast method. AV is very versatile plant, and its benefits are well known. MMT have gained popularity for internal and external detoxification. PVA is biodegradable material which is generally defined as the polymer which decomposes in the environment friendly atmosphere. The applicability of the evaluated formulations was influenced by the polyvinyl alcohol content due to their ability to alter the formulation viscosity. Their properties, relating mechanism and corresponding applications were deeply investigated. Significant formulation provided exclusive levels of applicability as well as thin-film forming performance, the minimum bare time possible and admirable homogeneity of the green-clay particles and aloe vera on or before dehydrating. Hence, the fundamental stability investigation which may optimize formulation is stable under minimal storage conditions. Furthermore, microbiological stability evaluation indicated that the preservative was efficient in term of avoiding microbial growth dynamics. Profitable statistical tools for the conviction of the nature of various compounds and their concentrations for the responses investigated, allowing the observation of the optimum situations further improvement of green clay and aloe vera peel-off skin masks.

Key words: Aloe vera; MMT; PVA; Polymeric membrane; Skin mask formulation

List of abbreviations

AV - Aloe vera

MMT - Montmorillonite

PVA - Poloy vinyle alcohol

XRD - X-ray diffraction

FTIR – Fourier transform infra-red

1. Introduction

Increasing demand of cosmetics all over the world from teen to adult has increased awareness related to safety issues. Nowadays the harmful effect of cheap chemical based cosmetics is a common problem. These chemicals are either added intentionally purpose or due to the manufacturing processes. Due to the reason biodegradable products are becoming more popular. Dermal exposure is expected to be the most significant route for cosmetic products, since the majority of cosmetics are applied to the skin [1]. According to the Dr. Hauschka, a leading pioneer in holistic skin care. “The average woman absorbs 30 pounds of the ingredients contained in moisturizers over sixty years” [2]. On the other hand organic and herbal products gently clean and moisturize the skin without causing dryness and are great for people with sensitive skin. These products contain many natural constituents that are easily absorbed by the skin keeping it smooth and healthy. Aloe vera soap and gel come loaded with several nutrients like glycerine, sodium palmate, sodium carbonate, sodium palm kemelate, sorbitol, etc. [3]. An aloe vera plant belongs to Liliacea family. Its leaves are full of parenchymatic tissues which contain 98.99% water and dry matters are made up if polysaccharides which is more than 60% [4]. The fresh gel or juice are formulated for general health, cosmetics and medical purposes but it is widely used as folk remedy [5]. These are good for the skin and nourish your skin from within, giving you the skin that glows with health. Wrinkles and fine lines are bound to appear as you age, but, other factors may expedite, and natural process can slow down. Aloe vera helps in preventing these early signs of aging. Aloe vera increases the water content in your skin, and leaves it hydrated without making it greasy. It contains 8 an enzyme in which bradykinase is useful for reducing excessive inflammation on topical application. Aloe vera has many minerals such as calcium,

potassium, zinc, sodium selenium and copper etc. which are very much essential for better functioning of enzymes and they are also antioxidants. From the mucilage layer of aloe vera plant many monosaccharides and polysaccharides are derived such as mannose -6-phosphate (monosaccharides) and beta - 4 - acetylated mannan/ glucomannans. Recently an anti-allergic glycoprotein known as alprogen and anti-inflammatory (C-glycosyl chromane) also derived from aloe Vera [6-7]. It has also wound healing hormones like auxins and gibberellins. This plant has 4 plant steroids i.e. cholesterol, campesterol, lupeol and beta - sitosterol with the properties of antiseptic and analgesic. It contains out of 22 amino acids it has 20 and out of 8 essential amino acids it has 8. Which are antibacterial in nature? Most importantly it has Lignin which enhances the absorbance of other substances in skin. In gel, 3% Saponins have property of cleansing and antiseptation [8]. Clay mineral is very much useful for the human life and civilization. It is an integral part of earth [9]. Today, the contribution of clay mineral is much significant in the field of health, additive and filler. Clay minerals are mainly used in dermatological clinics and spas. But in cosmetic industry these geoproducts are still a rising field that's why domestic markets are low [10-11]. Montmorillonite, is the one of the oldest clay, initially investigated in the region France so called 'Montmorillon' [12]. Clay stems back far in history as a traditional healing method for protecting the body from disease. Clay is readily available and it can be used in its original form without further processing. The use of MMT clay is a cheap and "detoxing" agent for human body for long time. It is very effective and safe way to get rid of toxins from body [13]. Clays are experimentally proven its traditional and natural utility for removing the skin irritation or other internal detox. [14]. MMT clay is a fine grained structure with sheet like geometry with best swelling behaviour having the absorption of solvent molecules in the clay galleries and also the adsorption of solvent molecules on its external sheet like surfaces [15]. Poly (vinyl alcohol) [PVA] is acknowledged to be one of few vinyl polymers that can have swelling behaviour with higher

biodegradation rate. This is possible owing to the presence of hydroxyl groups which condition hydrophilic nature of this material [16]. PVA has been frequently used in the preparation of various membranes and hydrogels [17-18]. Due to very low modulus of elasticity PVA gels causes minimal mechanical irritations and also shows very good biocompatibility with blood, body fluids and tissues [19]. These interpenetrating PVA hydrogels are formed due to swelling of monomers and the later reactions forms intermeshing network structure [20]. Additionally, the hydroxyl (-OH) groups, PVA is strongly hydrophilic and soluble in water, which helps its degradation through hydrolysis. Due to its biodegradable nature, PVA nanocomposites and their cross linked products have been widely investigated [21]. Aloe vera also used to prevent radiation-induced skin reactions [22]. Many of the health benefits associated with Aloe vera have been attributed to the polysaccharides contained in the gel of the leaves [23]. In this manuscript, we report Poly vinyl alcohol (PVA), Aloe Vera (AV) and Montmorillonite (MMT) with hydrated Base and an attempt is made to prepare gel $([0.4\text{PVA}-0.6\text{AV}]_{0.98}-[\text{MMT}]_{0.02})$ for the application of moisture and peeling off study. The physical property of the synthesised gel was studied by XRD and FT-IR spectroscopy. For the manual removal of the mask without leaving any residue create a very cohesive plastic layer after complete drying. This peel off facial mask is known for their unique characteristics and firming action of these mask formulation gives a clear skin sensation [19, 24-25]. Even though there are some promising results of green nanomaterials [26], clinical effectiveness of oral or topical aloe vera is not sufficiently defined at present [R27].

2. Materials and methodology:

To develop the $([0.4\text{PVA}-0.6\text{AV}]_{0.98}-[\text{MMT}]_{0.02})$ gel we used the following systematic process as follows;

2.1 Plant material

The Aloe vera (*Aloe Barbadensis* Miller) leaves were collected and processed from a single garden plant of our local area and authenticated by the *Department of Botany, Faculty of Science, University of Allahabad, India*.

2.2 Chemicals

Polyvinyl alcohol (PVA) polymer of 98-99% hydrolyzed (molecular weight, M.W. =98,000 g/mol, Across organics). MMT clay (Monmorillonite) is purchased from Sigma Aldrich. It has nanosize and layered structure. All samples and solution were prepared with water purified by double distillation process.

2.3 Peel- off mask Gel formulation from Aloe vera leaves

For the best film forming developed gel was mixed with water and was optimised to prepare peel off gel based on its consistency (Table-1)

Table 1. Optimized concentration of the chemical ingredients for the peel off gel formulation for skin mask

Chemical compounds	Categories	Optimized Concentration (%)
Aloevera (AV)	Gelling agent	6
Poly Vinyl Alcohol (PVA)	Film former	4
Monmorillonite (MMT)	Clay powder	0.2
Water	Base	~90

In this process, PVA was dissolved in doubled distilled water, at T=40°C (using water bath) and extracted aloe vera gel mixed in this solution. Finally nano MMT clay was dispersed in PVA-AV solution. The solution was stirred for 4-6 hours for homogenous mixing. This gel solution is physically stable up to 30-40 days. To extend the preservation time we can use

small amount of preservative like methyl paraban or propyl paraban. Stepwise bio-chemical synthesis of Gel ($[0.4\text{PVA}-0.6\text{AV}]_{0.98}:[\text{MMT}]_{0.02}$) were prepared through solution cast technique as depicted in the following **Figure 1**. A special chemical structure illustrating in this study biochemical calcination of Aloe vera latex extraction was utilized as an efficient controlling agent of size and shape dependent morphology. The four major C-glycosyl constituents in the Aloe vera latex are aloin A, Aloin B, aloesin, and aloresin A, illustrated.

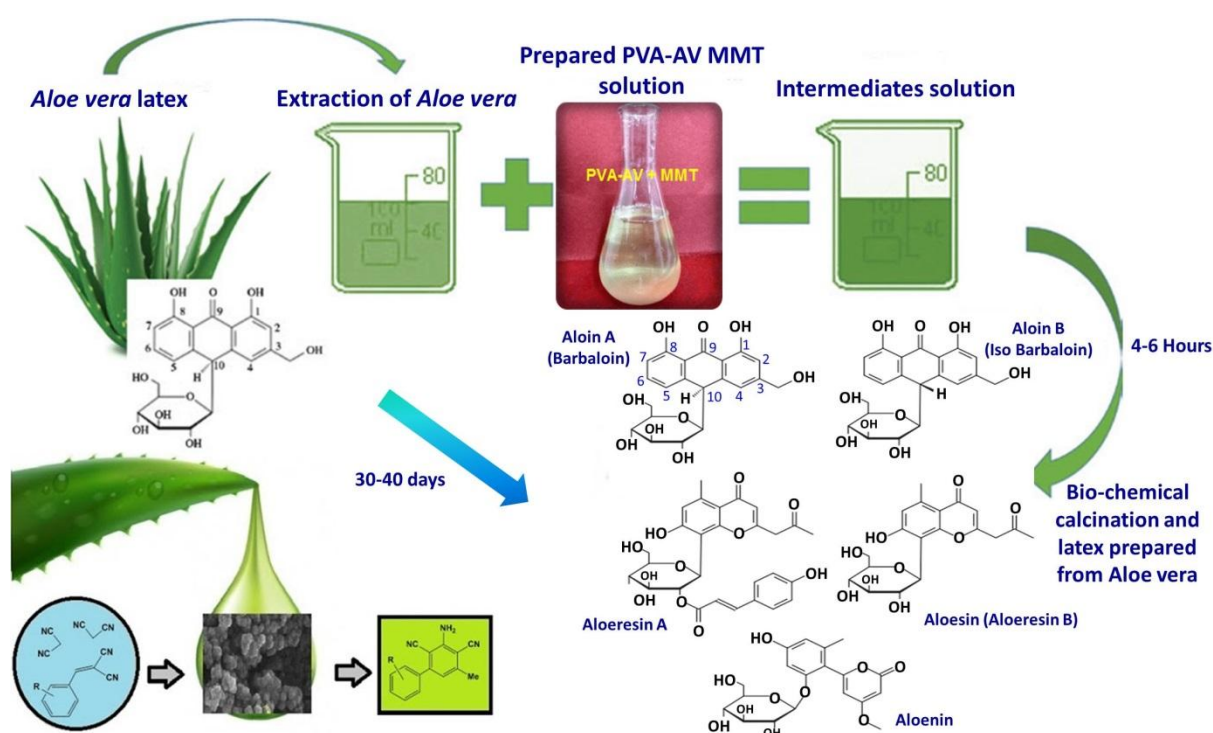


Figure 1. Eco friendly, excellent yields of the product, short reaction time, inexpensive, and readily available latex of *Aloe vera* material extracted prepared Gel ($[0.4\text{PVA}-0.6\text{AV}]_{0.98}:[\text{MMT}]_{0.02}$) in conical flask

The XRD pattern of the pristine and applied gel system was recorded using Phillips X-pert diffractometer in the 2θ range 10° to 75° . The infrared spectrum was recorded on Alpha ATR-FTIR Bruker (Germany) spectrophotometer in a range $4000-400\text{cm}^{-1}$.

3. Results and discussions

3.1 Application and characterization as peel off system

(i) *Optimized skin mask formulation*

Human body is the best creativity of the nature. But it is very sensitive to free radicals originated from the different sources of pollutants. To protect such harmful free radical use of face mask is a better option. The face mask is directly applied on the skin / face of human. Usually synthetic or chemical based gel packs dry the skin/face and produce harmful effect. The aloe vera based biodegradable product provides a better and healthy option. To study the moisturizing effect of gel system, applied on the back side of the bare palm and spread like film. After the application of gel the humidity of surface increases with respect to bare surface. It means the peel off gel mask from aloe vera gel has high emollient effectiveness. Other useful application is to clean the dirty and tired face after the attack of pollutant. This face mask cleanses, by smearing the gel directly onto skin surface having dirt, blemishes, red spots, irritations or scarring. Allow the gel to dry (Drying time 10-15 minutes) and peel off. We have used PVA polymer as base or film forming agent for peel off gel which is biodegradable water washable. Aloe vera is the important ingredient of this composite gel system. From ancient time, it was used for the cosmetic antithreptic application. Nowadays, it is become a fundamental active compound for the modern cosmology. It has also benefits in sunburn, bad case of acne, prevents signs of aging, moisturizes skin and also reduces marks. Aloe vera improves the elasticity of skin and makes it smoother and more supple [24]. Aloe vera leaves has two component, yellow juice has high content of 1,8-dihydroxy anthraquinone used for cathartic effect and mucilaginous gel is used for topical treatment [25]. It has gibberellins and auxins which reduces acne. It also contains polysaccharides that stimulate the growth of new cells [26]. On the basis of experimental results it's good to use the gel mask one or twice per week for best results. The application of synthesized peel off gel ($[0.4\text{PVA}-0.6\text{AV}]_{0.98}:[\text{MMT}]_{0.02}$) system and its mechanism is shown in **Figure 2**. While, in the **Figure 2(a)** shows that gel formulation were visually homogenous while still wet and

after drying formulation loses the homogeneity of the clay particle on the dried surface due to highly hydrophilic nature of PVA which aggregate during drying process and clay particle are dragged which is clearly shown in **Figure 2(b)**. Also, **Figure 2 (c)** explains the perforation and irregular thickness of membrane. Although drying time of ethyl alcohol higher than purified water but we have used purified water due to completely natural formulation. We have compared the drying time, applicability, film forming and desirability of the mask formulation. **Figure 2(d-g)** shows all the factors simultaneously. Addition of purified water/ethyl alcohol more than recommended for drying time in formulation would no longer be proportional (**Figure 2(d)**). The mask formulation has ideal applicability, so, low concentration of PVA has high level of applicability (**Figure 2(e)**). Hence, **Figure 2(f)** shows that PVA has certain concentration which is recommended to film forming performance. Increasing or decreasing is not beneficial for PVA uniform membrane and thickness. Desirability is an objective function and it completely depends on upper and lower limits. **Figure 2(g)** shows combined desirability function of formulation mask. In **Figure 2(h)** explains the applying process of formulation gel and peeling off mask.

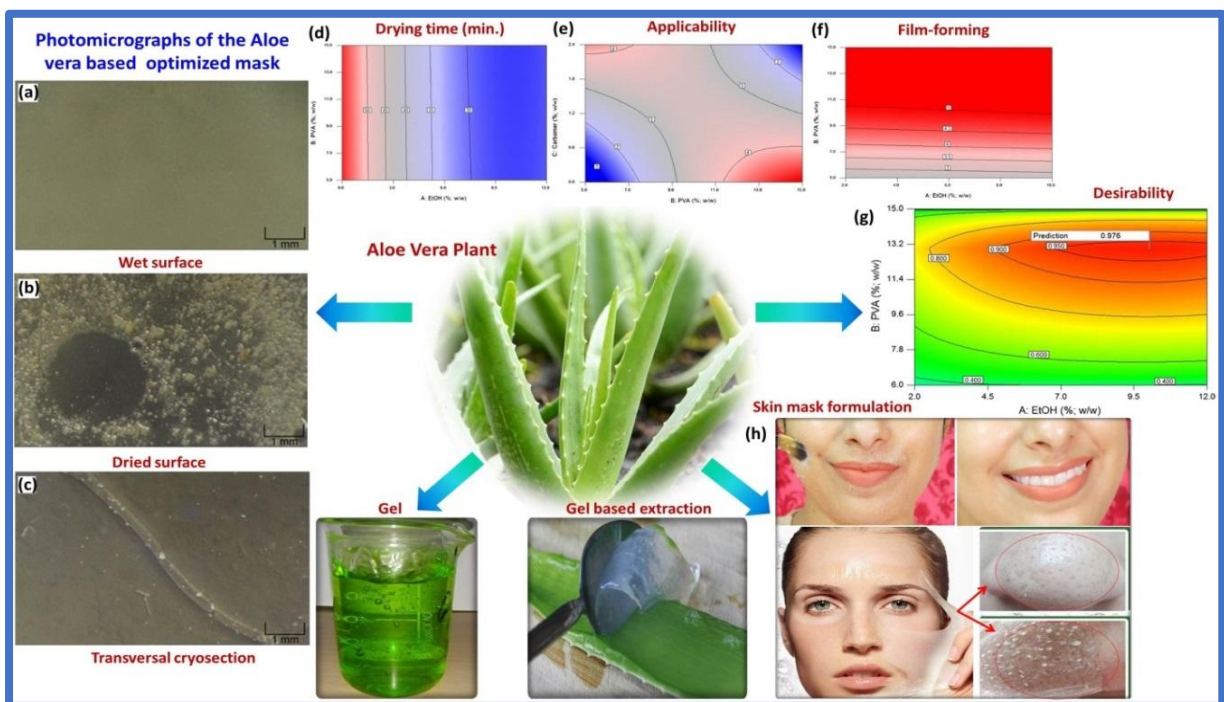


Figure 2.(a-c) Photomicrograph of the wet, dried and transversal cry section gel film (d-f) contour graph of drying time, applicability and film forming performance, (g) contour graphic of desirability of peel-off mask formulation (h) skin mask of formulation

This gel has anti-inflammatory properties to reduce sunburn caused by exposure to Ultra-violet vis. rays [27]. It acts on the epithelial skin layer and provides a protective layer to the skin for retaining moisture. Prepared gel has one component which is softening agent of gel i.e. MMT clay. Combination of aloe vera (hydrated) gel with MMT clay is able to bind free radicals, bacteria and toxins living on the surface of the skin and within pores to extract these from the pores and provide more moisturization. This helps to reduce the outbreak of blemishes, alleviate redness, and also to fight allergic reactions from irritating lotions or face washes [28]. MMT clay has the ability to produce a free charge that is electrical in nature when it comes in contact with liquid it starts works. When the clay touches any type of fluid (normally water), it takes on a different charge and is thought to bind to any present toxins within the fluid. Monmorillonite clay essentially “seeks” toxins in the body to bind with because naturally any substance that has a missing ion (which gives it its “charge”) looks for oppositely charged types of substances that will make it complete. Upon binding, the clay is then able to help remove toxins, chemicals, impurities and “heavy metals” from the gut, skin and mouth. To confirm the above property of Peeling/cleaning of the skin surface we tested the gel on two different artificial dirt surfaces. First one is has petroleum jelly type surface and second one has petroleum oil dust base surface. It can be easily noticeable colour difference of cleaned excrement skin and bare skin is given.

Table 2. Investigation of the optimized formulation for skin care prediction

	A: EtOH (%; w/w)	B: PVA (%; w/w)	C: Carbomer (%; w/w)	Drying time (min)	Applicability	Film forming
Predicted formulation	10.0	13.0	0	27	4.5	5.5
Confirmation runs (n = 3)	10.0	13.0	0	25	5	5
Individual desirability	1	0.9997	0.9999	0.9216	0.9806	1
Combined desirability	0.976	---	---	---	---	---
95% Confidence interval (CI)	---	---	---	24.6–30.8	3.7–6.4	4.7–6.1
95% Prediction interval (PI)	---	---	---	21.7–33.7	3.0–7.9	4.1–6.8

(ii) Mechanisms of skin mask formulation.

This gel has healing properties it accelerates the wound contraction and increases the collagen cross linking degrees [29]. Polysaccharides and gibberellins hormones stimulate the activity of fibroblast and also its proliferation which enhance the synthesis of collagen [30]. Skin mask has an antioxidants protein, metallothianien which scavenges the hydroxyl radicals from the skin and forms a protective layer against radiation [31-32]. This peel-off has ability to binds moisture to skin due to presence of many mucopolysacchrides in it. Amino acids decreases appearance of fine wrinkles by softening of skin and Zinc tightens the pores [33]. Due to presence of anti-inflammatory compound, C-glicosyle chromane in the gel, peel off mask inhibits the cyclooxygenase pathways [34]. This mask contains many antiseptic agents such as cinnamonic acid, sulphur, lupeol and salicylic acid which work against fungal and bacterial skin infections as shown in **Figure 3**.

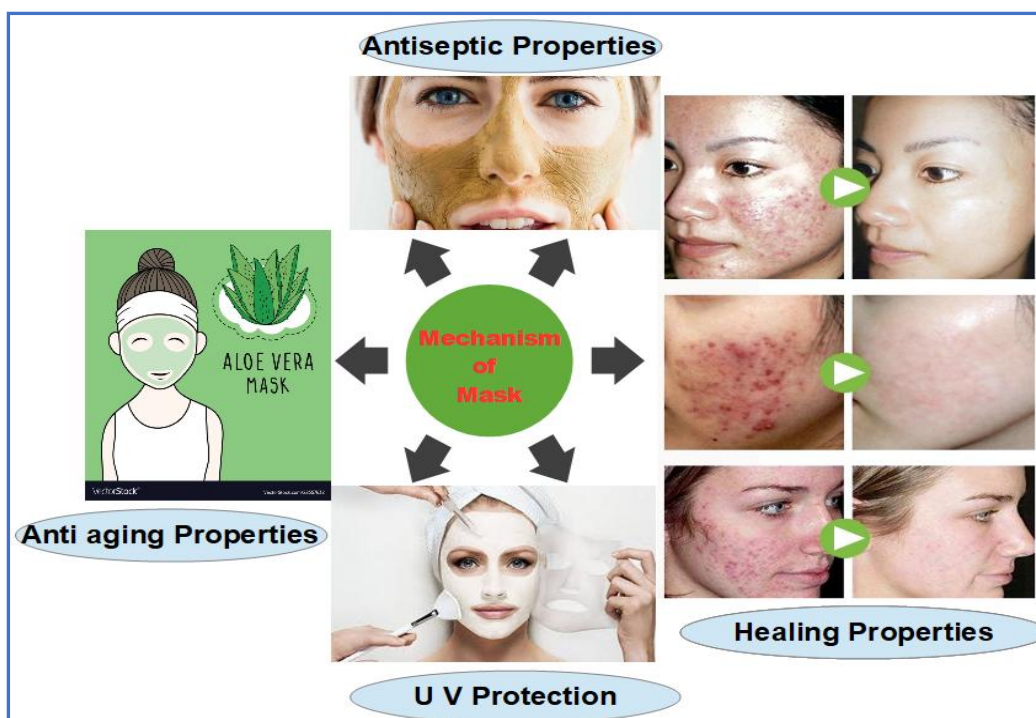


Figure 3. Schematic representation of peel-off mask mechanism

(iii) X-ray diffraction studies:

Typical X-ray diffraction pattern signifies polyvinyl alcohol and aloe vera solution of 40% PVA and 60% AV concentration in **Figure 4(a)**, while $[40\% \text{ PVA and } 60\% \text{ AV}]_{0.98} : [\text{MMT}]_{0.02}$ in **Figure 4(b)**, and $[40\% \text{ PVA-60\% AV}]_{0.98} : [\text{MMT}]_{0.02}$ with dust content (peeled-off) is shown in **Figure 4(c)**. Comparative study of $([40\% \text{ PVA} - 60\% \text{ AV}]$ (**Figure 4(a)**) and $[40\% \text{ PVA- } 60\% \text{ AV}]_{0.98} : [\text{MMT}]_{0.02}$) XRD pattern shows the amorphous nature of the gel, usually the polymeric system show amorphous nature. Addition of AV and MMT also enhance the broadening of the characteristics peak. And no individuals peak related to these constituents are present in the spectra (**Figure 4(b)**). This is indication of dissolution of AV and MMT in the film forming polymeric medium. The enhancements of peak area show the more amorphous or better homogeneity of the gel system. This gel was applied on the back of the palm surface and peel off after 20min. The peeling off product was also analysed by the X-ray diffraction technique. The XRD spectrum of final product is shown in **Figure 4(c)**. This

spectrum (19.86°) is become more crystalline in nature with respect to the virgin gel product. The sharpness of the peak indicates there is some crystallite particles i.e. dust particle spread on the skin. These particles are absorbed by the gel film putting on the skin. The second small broad peak (40.38°) is an indication of presence of some crude oil product which is present in the pollutant air of the atmosphere.

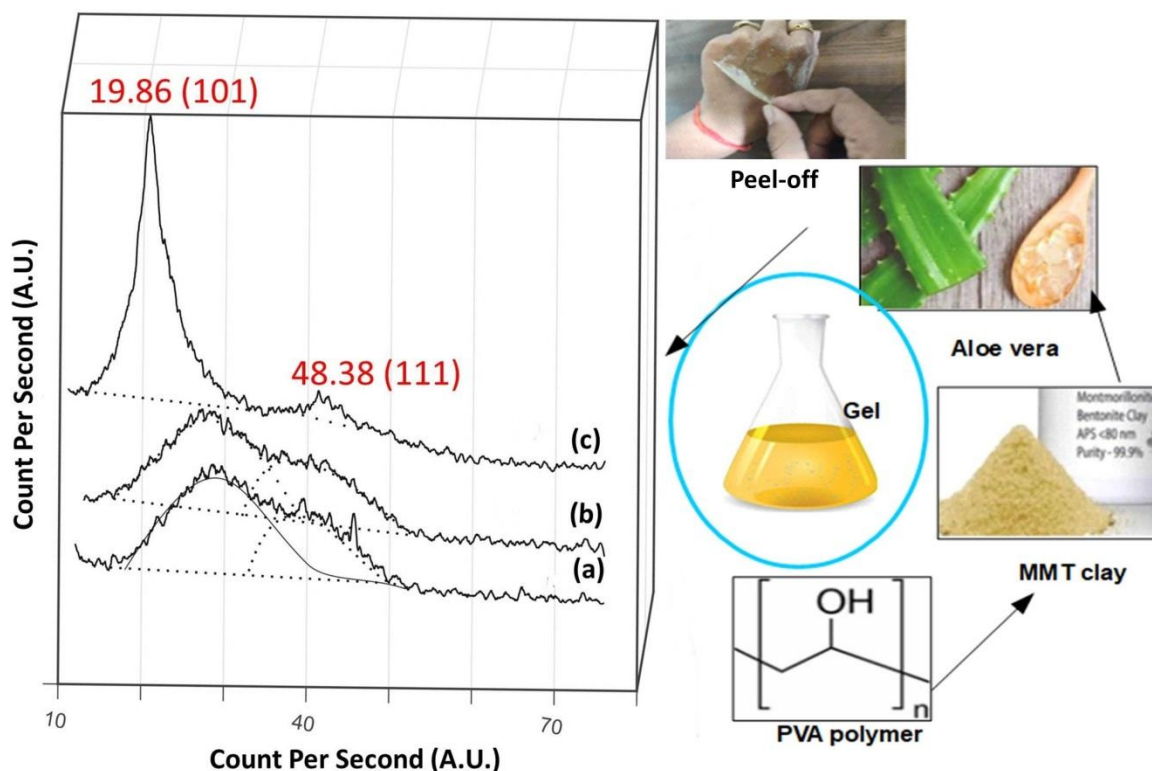


Figure 4. X-ray diffraction spectrum of (a) [40%PVA-60%AV], (b) [40%PVA-60%AV]_{0.98}: [MMT]_{0.02}, and (c) [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} with oil dust (peel-off) composite

(iv) FT-IR study of Peel- off gel membrane analysis

Our significant effort to investigate functional groups present in *Aloe vera* buds by using FT-IR spectroscopic technique which is simple, cost effective and user friendly technique to investigate functional groups of compounds in sample. FT-IR spectra of [40% PVA-60%AV] and [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} gel systems are shown in **Figure 5**. A comparative study of the both system shows the appearance of some prominent peaks of MMT based

system. These peaks are at 2953 cm^{-1} , 2910 cm^{-1} and 2849.4 cm^{-1} related to the characteristics signal of C-H Stretching of MMT. **Figure 6**, shows the FT-IR spectra of $[40\%PVA-60\%AV]_{0.98}:[MMT]_{0.02}$ gel and $[40\%PVA-60\%AV]_{0.98}:[MMT]_{0.02}$ with oil dust and petroleum gelly (peel-off) composite gel system. Petroleum gelly and oil dust peeled off membrane has C-C stretching at 1717.0 cm^{-1} and 1741.9 cm^{-1} due to bending of aromatic and aliphatic structures. The prominent peaks at 2843.4 cm^{-1} , 2921.9 cm^{-1} and 2953.6 cm^{-1} of MMT which present in host gel and also become more sharp and intense due to presence of crude oil dust and petroleum jelly which converts the gel in more crystalline nature. This is due to C-H anti-symmetric and symmetric stretching in an aliphatic hydrocarbon. More crystalline nature of both peeled off membranes is due to adsorption nature of dust and cosmetics from skin, which the characteristics feature of MMT. The FT-IR is to investigate number and types of bioactive compounds present in Aloe Vera buds grown in North East India at Allahabad region is taken, non-destructive, cost effective and user friendly. Aloe vera was cultivated under normal atmospheric conditions. FT-IR suggested that the presence of acetyl groups is necessary for biological activation, possibly because they cover a number of hydrophilic hydroxyl groups and thus make the molecule more able to cross hydrophobic barriers in the cell.

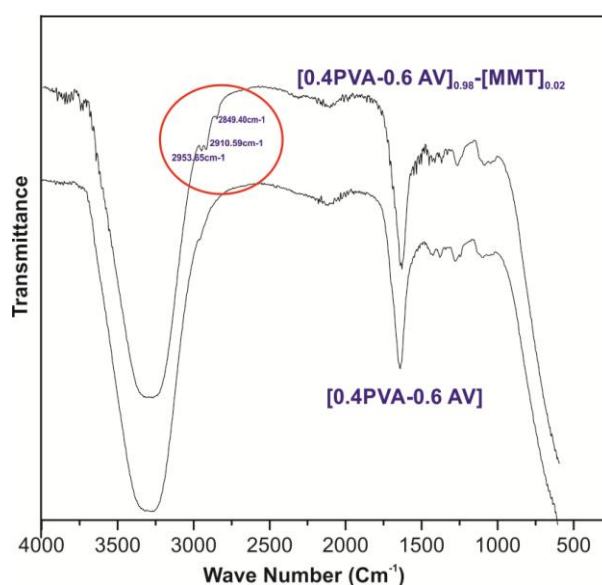


Figure 5. FT-IR spectra of [40%PVA-60%AV] and [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} composite

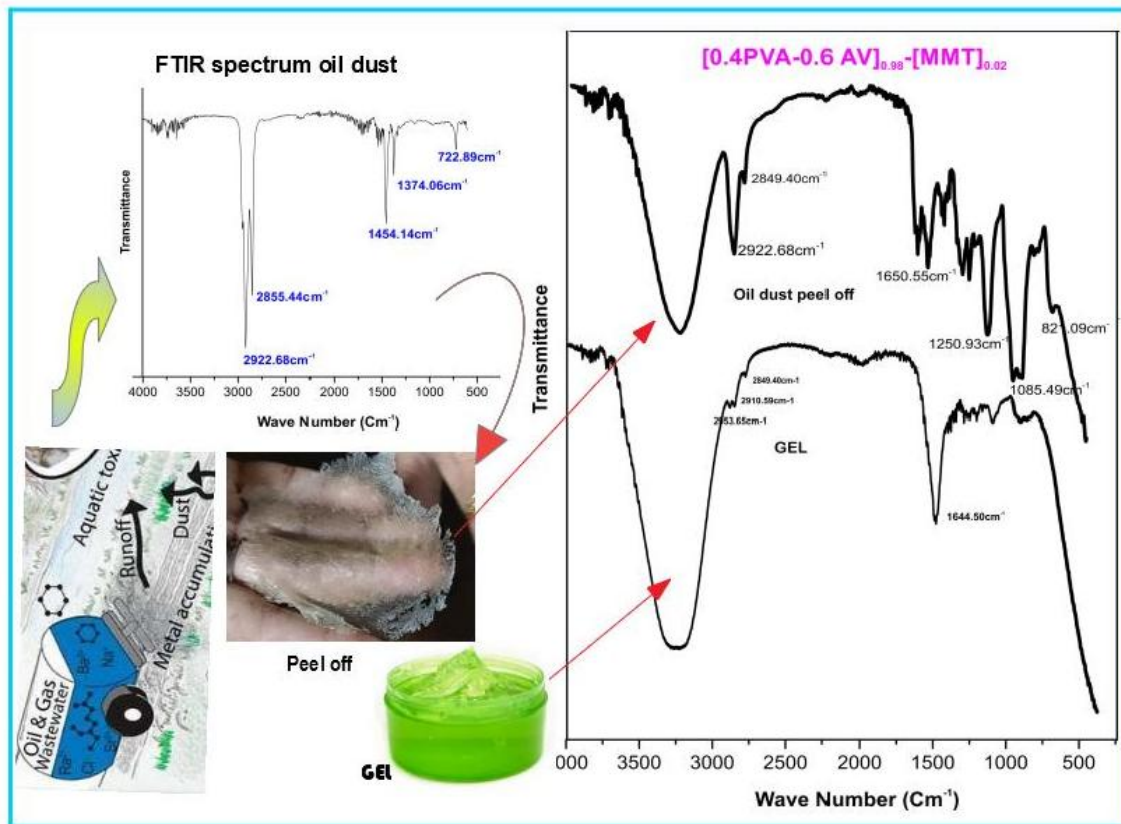


Figure 6. FT-IR spectra of [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} Gel and [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} with oil dust & petroleum jelly (peeled off) composite

4. Conclusions and outlook

The outcome of this study is the development of Aloe Vera, MMT incorporated PVA formed gel possessing many beneficial treat. These gel have excellent antimicrobial activity. These gels when applied as a face mask than all dust are adsorbed which is characterized and explained. Based on above mentioned features, these gels could be used in a wide range of application in cosmetics field. The significance of this work lies in the use of aloe vera and MMT for its exploitation to the anti-inflammatory. This will be useful to those peoples who are suffering from enormous skin diseases. The response of surface treatment was found to be

an emerging statistical tool to determine their concentrations based on response investigation, allowing the observation of the optimal conditions to evince refined-particle size green clay and aloe vera peel of facial masks. Further evidences of clinical trial if may conclude that suggest that topical Aloe vera is effective as well as sufficiently powered research studies should be conducted to evaluate the effectiveness of currently utilized and novel therapies for the prevention, minimisation and management of radiation-induced skin reactions. Demand for aloe vera product in food industry is expected to grow with a fastest rate in the forecast period. Wide ranges of utilizations provides background formation on the potential application of sealed drinking water, low-calorie fruit juice, as well as peel-off formulation further opening a brand new scopes in pharmaceutical ingredients.

Acknowledgments.

The author, Dr. Nidhi Asthana is grateful to *Department of Science and Technology, Government of India* for awarding the WoS-A Project (SR/WOS-A/CS-114/2016). Dr. Kaushik Pal is grateful to his associate colleagues and Doctoral/Postdoctoral research scholars' consistent involvement at the *Federal University of Rio de Janeiro, Brazil*. Especially, Dr. Pal, owing to his sincere thanks to all scientific operators and members are gratefully acknowledged for providing excellence of the research foundation.

Compliance with ethical standard:

Declarations of interest.

All the authors declared there is no conflict of interest for the publication of this work.

Research involving human participation and/or animals. None

Informed consent. None

Ethical approval. This article does not contain any studies with animals or humans performed by any of the authors.

References

1. Zulaikha S, Ismaael S N S, Praveen S M. Hazardous ingredient in cosmetics and personnel care products and health concern : A review, Public Health Research 2015;5:7-15
2. Elisabeth S, dr. hauschka.com, US expert-advice, 1967
3. Vijayalakshmi D, Dhandapani R, Jayaveni S, Jithendra P, rose C, mandal A B. In Vitro anti inflammatory activity of aloe vera by down regulation of MMP-9 in peripheral blood mononuclear cells. J. Ethnopharmacol. 2012; 141; 542-546.
4. Simal S., Feminia, A., LIull, P., Bruno, E., J. Ethanopharmacol; 1996; 55; 69.
5. Chithra ,P; Sajith lal, C.B.; Gowri, C. ; J. Ethanopharmacol; 1998; 59; 179.
6. Ro JY, Lee B, Kim JY, Chung Y, Chung MH, Lee SK, et al. Inhibitory mechanism of aloe single component (Alprogen) on mediator release in guinea pig lung mast cells activated with specific antigenantibody reactions. J Pharmacol Exp Ther. 2000;292:114–21.
7. Hutter JA, Salmon M, Stavinoha WB, Satsangi N, Williams RF, Streeper RT, et al. Antiinflammatory C-glucosyl chromone from Aloe barbadensis. J Nat Prod. 1996;59:541–3.
8. Amar S., Resham V., and D G Saple, Aloe Vera: A Short Review, Indian J Dermatol. 2008; 53(4): 163–166.
9. Report on Health Hazards in Cosmetics product.
10. Carretero M.I., Lagaly, G.; clays an health ; an introduction, appl. Clay Sci ; 2009; 36; 1-

11. Ghersetich, I.; Lotti T.M., immunological aspects ; immunology of water spas. Clin. Dermatol.; 1996; 14; 563-566.
12. Uddin F. Clay nanoclay and montmorillonite minerals, Metallurgical and Materials Transaction A. December 2008; 39 : 2804-2814
13. Fowler J , L; W, Bailey C; Effects of a calcium Bentonite clay in Diets containing Aflatoxin when measuring Liver Residues of Aflatoxins B in starter Broiler chicks. Toxins (Basel); 2015;7; 3455-64
14. Maryam Moosavi; Bentonite Clay as a Natural Remedy: A Brief Review Iran J Public Health. 2017; 46: 1176–1183.
15. G. Lagaly, Introduction: from clay mineral-polymer interactions to clay mineral-polymer nanocomposites, Applied Clay Science, 1999; 15, 1–9.
16. Chandra, R. and Renu, R. Biodegradable Polymers. Progress in Polymer Science 1998; 23: 1273-1335
17. Hatice, B.; Nursel ,P.; Guven , O.; Radiant phys. Chem.. 1999; 55; 667
18. Zhai, M.; Ha H.; Yushi F.; Makuuchi K.; Radiant Phys. Chem. 2000; 57; 459
19. Rosiak, J.m.J.; controlled release, 1994; 31;9
20. Pappas, N.A.; Biomaterials science: an introduction to materials in medicine academics: San Diego. C.A. 1996
21. Tayser Sumer Gaaz , Abdul Amir H. Kadhum , Patina Kiah Anak Michael , Ahmed A. Al-Amiery , Abu Bakar Sulong , Mohamed H. Nassir and Ahed Hameed Jaaz; Unique Halloysite Nanotubes–Polyvinyl Alcohol–Polyvinylpyrrolidone Composite Complemented with Physico–Chemical Characterization; Polymers 2017, 9, 207-222
22. J. Richardson, J.E. Smith, M. McIntyre, R. Thomas, K. Pilkington, Clinical Oncology, 17(6), 2005, pp. 478-484
23. J. H. Hamman, *Molecules* 2008, 13(8), 1599-1616

24. De Naverre, M.G.; the chemistry and manufacture of cosmetics 2nd ed. Orlando; Continental; 1975
25. Baby,A.R.; Zaque, V.; Macial, C.p.M; Kaneko T.M.,; Consiglieri, V.C.; Valesco, M.V.R.; Development of cosmetic mask formulations. Rev. Bras Cienc Farm 2004;40; 159-161
26. Asiya SI, K Pal, S Kralj, GS El-Sayyad, FG de Souza, T Narayanan, Mat.Today Chem.,17(2020), 100327
27. B. K. Vogler, E. Ernst, *British Journal of General Practice* 1999; 49 (447), pp.823-828.
28. Davis RH. Biological activity of aloe vera. SOFW Journal; 1993; 119; 646–49.
29. Beatriz, V.; Guillermo, A.; David, S.; Bruno E.; J Ethnopharmacol 1996, 55,69.
30. Chithra P, Sajithlal GB, Chandrakasan G. Influence of aloe vera on collagen characteristic in healing dermal wound in rats. Mol Cell Biochem. 1998; 181:71–6.
31. Ito S, Teradaira R, Beppu H, Obata M, Nagatsu T, Fujita K, Properties and pharmacological activity of carboxypeptidase in Aloe arborescens Mill var. natalensis Berger, Phytother Res, 1993; 7; 26–9.
32. Shelley E H, Christine M R, And Lynda B W, Broad-spectrum in vitro antibacterial activities of clay minerals against antibiotic-susceptible and antibiotic-resistant bacteria pathogens, J Antimicrob Chemother, 2008 ; 61; 353–361.
33. Hegggers J, Kucukcelebi A, Listengarten D, Stabenau J, Ko F, Broemeling LD, et al. Beneficial effect of aloe on wound healing in an excisional wound model. J Altern Complement Med. 1996; 2: 271–277.
34. Chithra R Sajithlal GB, Chandrakasan G. Influence of aloe vera on collagen characteristics in healing dermal wounds in rats. Mol Cell Biochem. 1998; 181: 71–76.
35. Roberts DB, Travis EL. Acemannan-containing wound dressing gel reduces radiation-induced skin reactions in C3H mice. Int J Radiat Oncol Biol Phys. 1995; 32: 1047–1052.

36. Sato Y, Ohta S, Shinoda M. Studies on chemical protectors against radiation XXXI: Protective effects of *Aloe arborescens* on skin injury induced by x-irradiation. *Yakugaku Zasshi*. 1990; 110: 876–884.
37. West DP, Zhu YF. Evaluation of aloe vera gel gloves in the treatment of dry skin associated with occupational exposure. *Am J Infect Control*. 2003; 31: 40–42.
38. Shelton M. Aloe vera, its chemical and therapeutic properties. *Int J Dermatol*. 1991; 30: 679–683.

Research highlights

- Significant applicability as well as thin-film performance by green-clay particles and aloe vera
- PVA-based aloe vera gel was prepared via bio-chemical solution cast technique
- Optimized formulation is stable under the normal storage conditions
- Various diversities in peels-off facial mask firming action of a clear skin sensation

Figures file

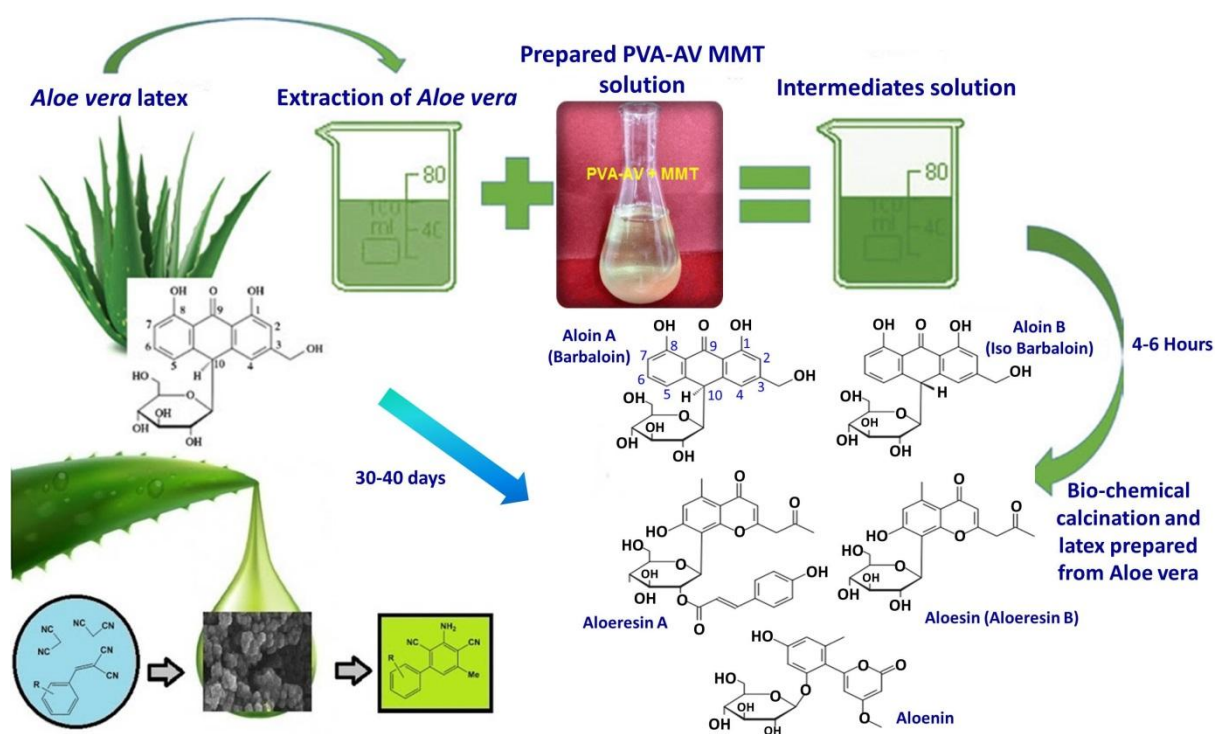


Figure 1. Eco friendly, excellent yields of the product, short reaction time, inexpensive, and readily available latex of *Aloe vera* material extracted prepared Gel ([40%PVA- 60%AV] 0.98:[MMT]_{0.02}) in conical flask

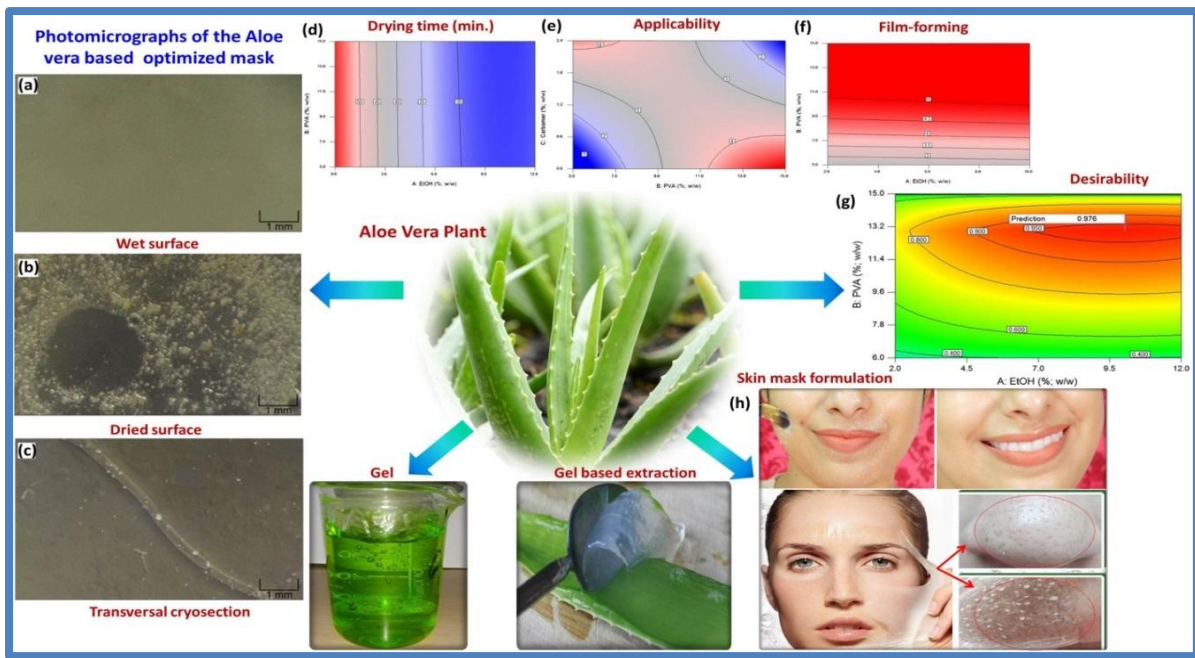


Figure 2.(a-c) Photomicrograph of the wet, dried and transversal cryosection gel film (d-f) contour graph of drying time, applicability and film forming performance, (g) contour graphic of desirability of peel-off mask formulation (h) skin mask of formulation

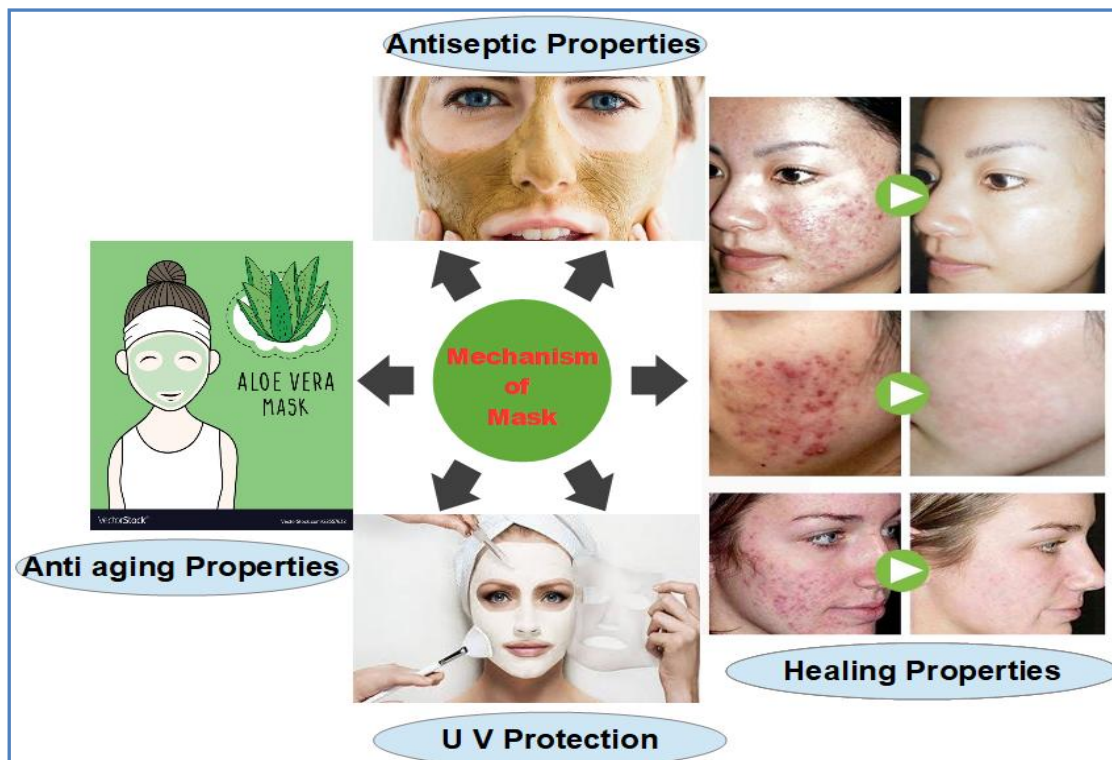


Figure 3. Schematic representation of peel-off mask mechanism

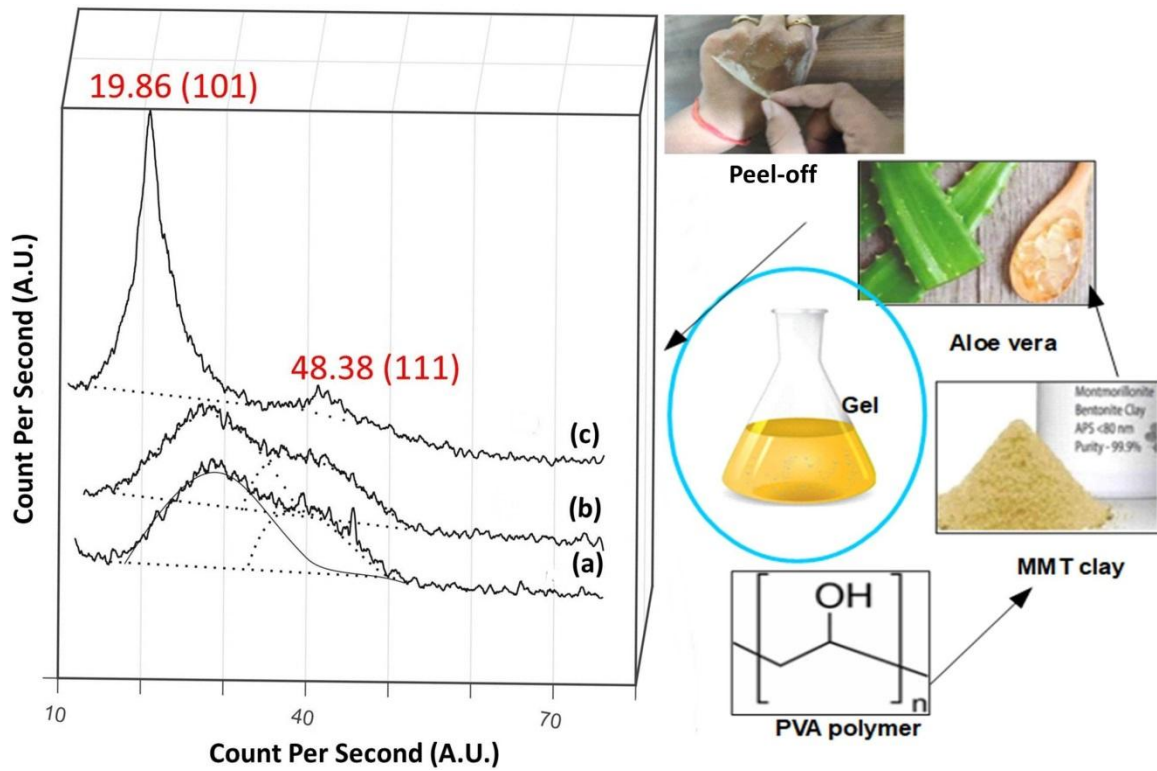


Figure 4. X-ray diffraction spectrum of (a) [40%PVA-60%AV] (b) [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} and (c) [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} with oil dust (peeled off) composite

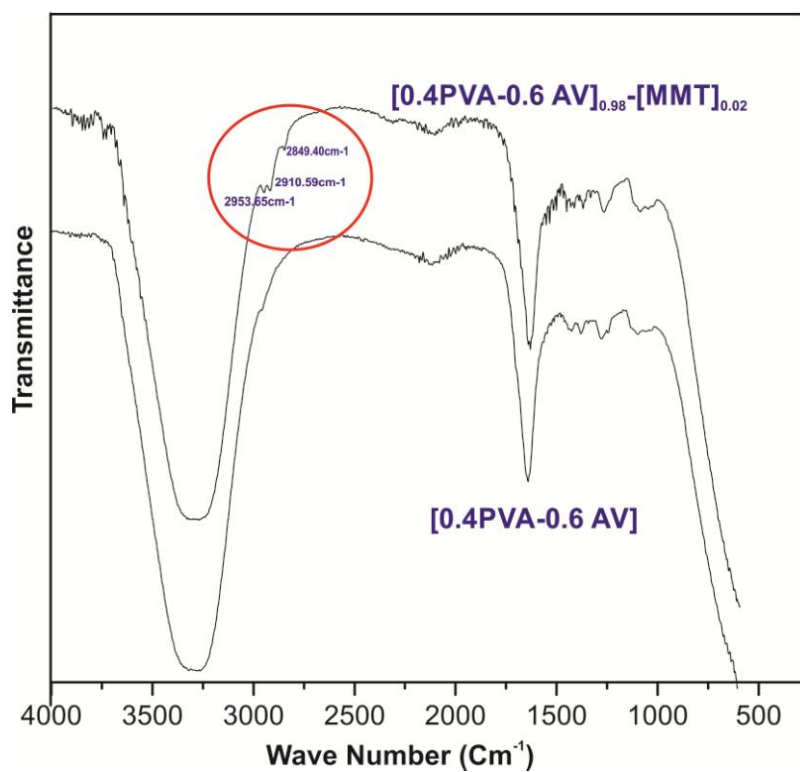


Figure 5. FT-IR spectra of [40%PVA-60%AV] and [40%PVA-60%AV]_{0.98}: [MMT]_{0.02} composite

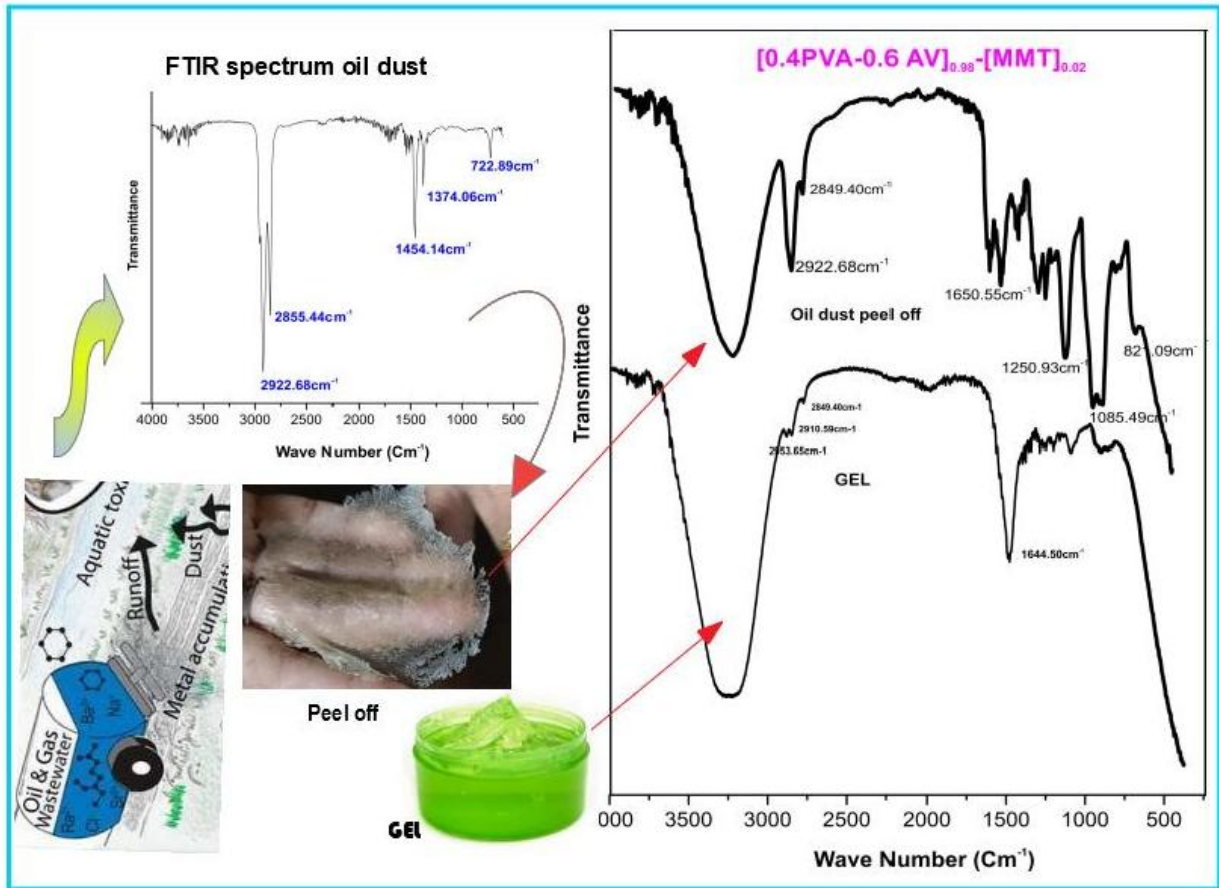


Figure 6. FT-IR spectra of $[40\% \text{ PVA}- 60\% \text{ AV}]_{0.98}:[\text{ MMT}]_{0.02}$ Gel and $[40\% \text{ PVA}- 60\% \text{ AV}]_{0.98}:[\text{MMT}]_{0.02}$ with oil dust & petroleum gelly (peeled off) composite

Table 1. Optimized concentration of the chemical ingredients for the peel off gel formulation for skin mask

Chemical compounds	Categories	Optimized Concentration (%)
Aloevera (AV)	Gelling agent	6
Poly Vinyl Alcohol (PVA)	Film former	4
Monmorillonite (MMT)	Clay powder	0.2
Water	Base	~90

Table 2. Investigation of the optimized formulation for skin care prediction

	A: EtOH (%; w/w)	B: PVA (%; w/w)	C: Carbomer (%; w/w)	Drying time (min)	Applicability	Film forming
Predicted formulation	10.0	13.0	0	27	4.5	5.5
Confirmation runs (n = 3)	10.0	13.0	0	25	5	5
Individual desirability	1	0.9997	0.9999	0.9216	0.9806	1
Combined desirability	0.976	---	---	---	---	---
95% Confidence interval (CI)	---	---	---	24.6–30.8	3.7–6.4	4.7–6.1
95% Prediction interval (PI)	---	---	---	21.7–33.7	3.0–7.9	4.1–6.8

Credit Authors Statement

All the authors have declared that there is no conflict of interest for the publication of the following manuscript **Title of the article:** Polyvinyl alcohol (PVA) mixed green–clay and aloe vera based polymeric membrane optimization: Peel-off mask formulation for skin care cosmeceuticals in green nanotechnology, in **Journal of Molecular Structure, (Elsevier)** special issue entitled ‘**Nanoarchitectonics: From molecules to Advanced nanomaterials**’

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Nidhi Asthana

Dr. NIDHI ASTHANA
Women Scientist (WOS-A DST)
National Centre Of Exp. Mineralogy and Petrology
14-Chatham Line Bank Road
UNIVERSITY OF ALLAHABAD
ALLAHABAD