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(RESEARCH ARTICLE)

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A novel approach to potentiate a Unani formulation – Marham Kafoori – using mid-infrared rays

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Abstract

Worldwide, unani, an age-old medical system with Greek roots, is still in use. Potentiation in Unani remedies needs to be redesigned to meet the needs of newly emerging ailments, however potentiation has not yet been tried in the Unani system. In this study, we developed a safer and more cost-effective way to apply and potentiate Marham Kafoori, a Unani medication used for topical wounds, using a mid-infrared generating atomizer (MIRGA). With a 20–40% shorter treatment duration, the potentiated Unani medication was found to be more effective. Clinical dose reduction, host stress reduction, and resource and expense reductions follow. Using FTIR and NMR spectroscopy, MIRGA-induced modifications in Marham Kafoori were further examined in terms of chemical bond configurations and alterations, and the results are described in full below.

Keywords: MIRGAl; 2-6 µm mid IR; Unani drug; *Marham Kafoori;* Potentiation; Resource; Economic savings; Safe.

1. Introduction

For millennia, the ancient medical system known as unani medicine has been used. The WHO has acknowledged Unani as an alternative system to cater our health care needs (Hongal et al., 2014). It is necessary to modify and intensify Unani formulas in order to cure modern ailments. However, the high-potency Unani formulations are now toxic by nature, and in order to render them suitable for therapeutic use, they must go through several processes like detoxification. Additionally, evaluating and validating the revised Unani medications using the current parameters and putting them into practice would take time (Chandra, 2017). Therefore, rather than creating a new formulation, we used mid-infrared energy to safely potentiate the existing formulation, in an easy and economical way without adverse effects.

2. Material and Method

MIRGA *(patent no.: 401387)* is a 20-mL capacity polypropylene plastic atomizer/ sprayer containing an inorganic (molar mass 118.44 g/mole) water-based solution. The sprayer unit has dimensions 86 × 55 × 11 mm, an orifice diameter of 0.375 mm, ejection volume 0.062 ± 0.005 mL, and ejection time 0.2 s. The average pressure is 3900 Pa, and the cone liquid back pressure is 2000 N/m^2. Design of the MIRGA and emission of 2-6µm mid-IR has been presented in detail by Umakanthan *et al*., 2022a; Umakanthan *et al*., 2022b. *(Fig 1)*

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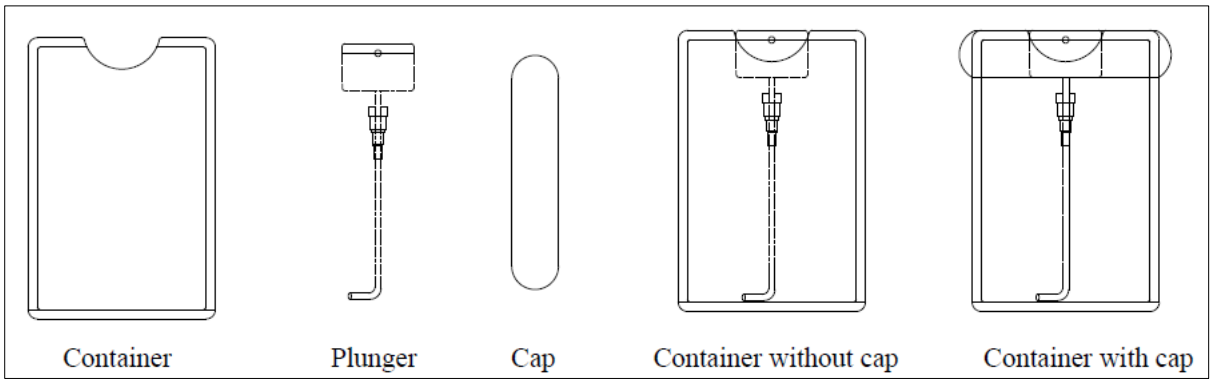

Figure 1 MIRGA sprayer

During spraying, approximately 1-µg weight of water is lost as mist and the non-volatile material in the sprayed liquid has a concentration of 153 mg/mL. Every time spraying emits 0.06ml which contains approximately seven quintillion cations and eleven quintillion anions. Depending on the pressure applied to the plunger, every spraying is designed to generate 2–6 µm as estimated by an FTIR (retro-reflector) interferometer instrument (Detector type D* [cm HZ1/2 - 1] MCT [2-TE cooled]) at Lightwind, Petaluma, CA, USA.

Spraying was performed at a distance of 0.25 to 0.50 meter from polythene packaged *Marham Kafoori*. This distance is needed for the MIRGA-sprayed solution to form ion clouds, oscillation and 2-6 µm mid-IR generation *(dealt in Discussion section)*. The mid-IR penetrated the intervening polythene package material (polythene/ glass) and act on the *Marham Kafoori* inside. Spraying at a close distance does not generate energy. MIRGA used like a body spray externally over packed *Marham Kafoori*, or other Unani formulations.

Method of MIRGA spraying: https://drive.google.com/open?id=1QoRwTESKfSdoJTfD--xIG9YpTDnVonGW

Marketed *Marham Kafoori* ointment – an Unani herbal medicine containing Kafoor as chief ingredient – was tested. Domestic animals (12 cattle and 12 dogs) were employed. Twenty packets each of 50g *Marham Kafoori was purchased.* The 20 packets were divided into 4 batches with 5 in each. First batch received no MIRGA spraying which formed as control. Second, third and fourth batches respectively received 2, 4, and 6 number of MIRGA sprayings, from a distance of 0.25 to 0.50 meter.

Four groups I, II, III and IV each containing randomly selected 3 cattle and 3 dogs of either sex, different ages and suffering from topical wounds were assigned, in total 24 animals. Control, 2, 4 and 6 sprayed *Marham Kafoori were respectively applied to the animals of group I, II, III and IV,* twice daily over the wounds until healing. The efficacy of the control and sprayed *Marham Kafoori was* assessed based on the number of days taken for wound healing and compared across groups. Sample from control, 2, 4 and 6 sprayed *Marham Kafoori were* also subjected to GCMS, FTIR and 1H-NMR to identify the changes caused by MIRGA spraying.

Ethical approval: In this study, it was only the *Marham Kafoori* ointment powder subjected to mid-infrared spraying externally over the packets. When we approached the institutions for ethical approval, they quoted the above reason and also because the study did not directly involve the application of mid-infrared application over the human/ animal patients, this won't come under the purview of ethical approval and so they refused our request. No ethical approval sought.

We used an increased number of sprayings (i.e. 6 sprayings) because, in nature, the input of more energy to any medicine should denature the medicine's natural characters and reduces its efficacy. Hence we tried this phenomenon also by 6 times spraying the Unani formulation until denatured.

The instruments used to identify the internal effect of mid-IR on *Marham Kafoori are*:

Gas chromatography mass spectrometry (GC-MS) for chemical compound transformation: Agilent 7890A GC with 5975C MS system. Column: HP-5. Ionization: EI (70 eV). Method: General_1_HP5_80_DEG.M. MSD: Single Quad

Fourier-transform infrared spectroscopy (FTIR) for chemical bond changes: JASCO FT-IR 4200 plus spectrophotometer with ATR (range 4000–400 cm^{-1} at 298 K)

Nuclear magnetic resonance spectroscopy (NMR) for proton resonances: Bruker AV-500 high resolution multinuclear FT-NMR spectrometer equipped with a 5 mm rotor was used for 1H and 13C measurements. Number of scans: varied from 8 to 64 for 1H measurements and 256-1024 for 13 C measurements. CDCl3 or DMSO -d6 were used according to the samples solubility. Bruker TOPSPIN software is used for integration and chemical shift calculation.

3. Results and Discussion

Time taken for complete healing of wound(s) was: 5-10 days in animals which received the control/ non-sprayed formulation. Whereas it was 5-7 days with 2 sprayed, 4-6 days with 4 sprayed, and 7-11 days with 6 sprayed *Marham Kafoori*. *(Fig 2)*

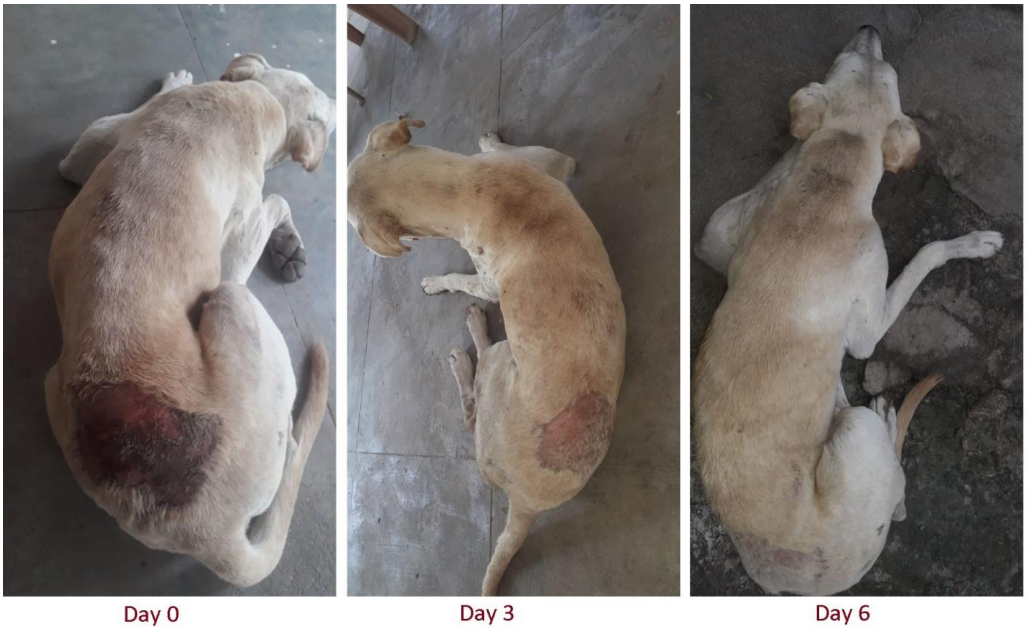

Figure 2 Effect of sprayed Markam Kafoori on abrasion wound in a dog

4. Instrumentation results

4.1. Raw data files of instrumentations

https://drive.google.com/open?id=1Hmz49shlHBEQ0nxykdmCRx-a85CYiiQw

4.1.1. GC-MS

Table 1 GC-MS spectral peak analysis

RT (Min)	Name of compound	% area present in each sample				Remarks
		Control	2 sprayed	4 sprayed	6 sprayed	
10.659	Bicyclo[2.2.1]heptan-2-one, 1,7,7-trimethyl-, (1S)-	6.789	11.268	11.860	16.912	Most abundant peak in 6 sprayed sample
42.286	2-Hexyl-1-octanol	4.869	3.165	2.183	3.395	
44.939	Tetracontane, 3,5,24-trimethyl-	5.620	4.721	5.845	4.626	
47.500	1-Iodo-2-methylundecane	12.209	11.111	7.610	7.564	
49.976	Eicosane	6.590	9.624	7.412	8.294	

52.407	Octadecane, 2-methyl-	9.673	11.527	6.744	9.154	Most abundant peak in 2 sprayed sample
54.833	Octadecane, 2-methyl-	6.829	7.707	8.807	9.900	
56.497	Octadecane, 6-methyl-	7.259	5.324	6.216	9.463	
57.768	Octadecane, 6-methyl-	5.977	7.815	7.222	7.611	
58.857	Heptadecane, 9-hexyl-	5.872	8.514	6.116	7.633	
59.542	Ethyl iso-allocholate	0.0	10.129	5.756	7.633	
60.020	Eicosane	0.0	6.132	9.357	7.042	
60.573	Hexadecanoic acid, 1-(hydroxymethyl)-1,2-ethanediyl ester	24.809	0.0	0.0	0.0	Most abundant peak in control
61.291	Ergosta-5,22-dien-3-ol, acetate, (3β,22E)-	3.502	0.0	0.0	0.0	
61.327	Ethanol, 2-(octadecyloxy)-	0.0	2.963	14.873	3.245	Most abundant peak in 4 sprayed sample

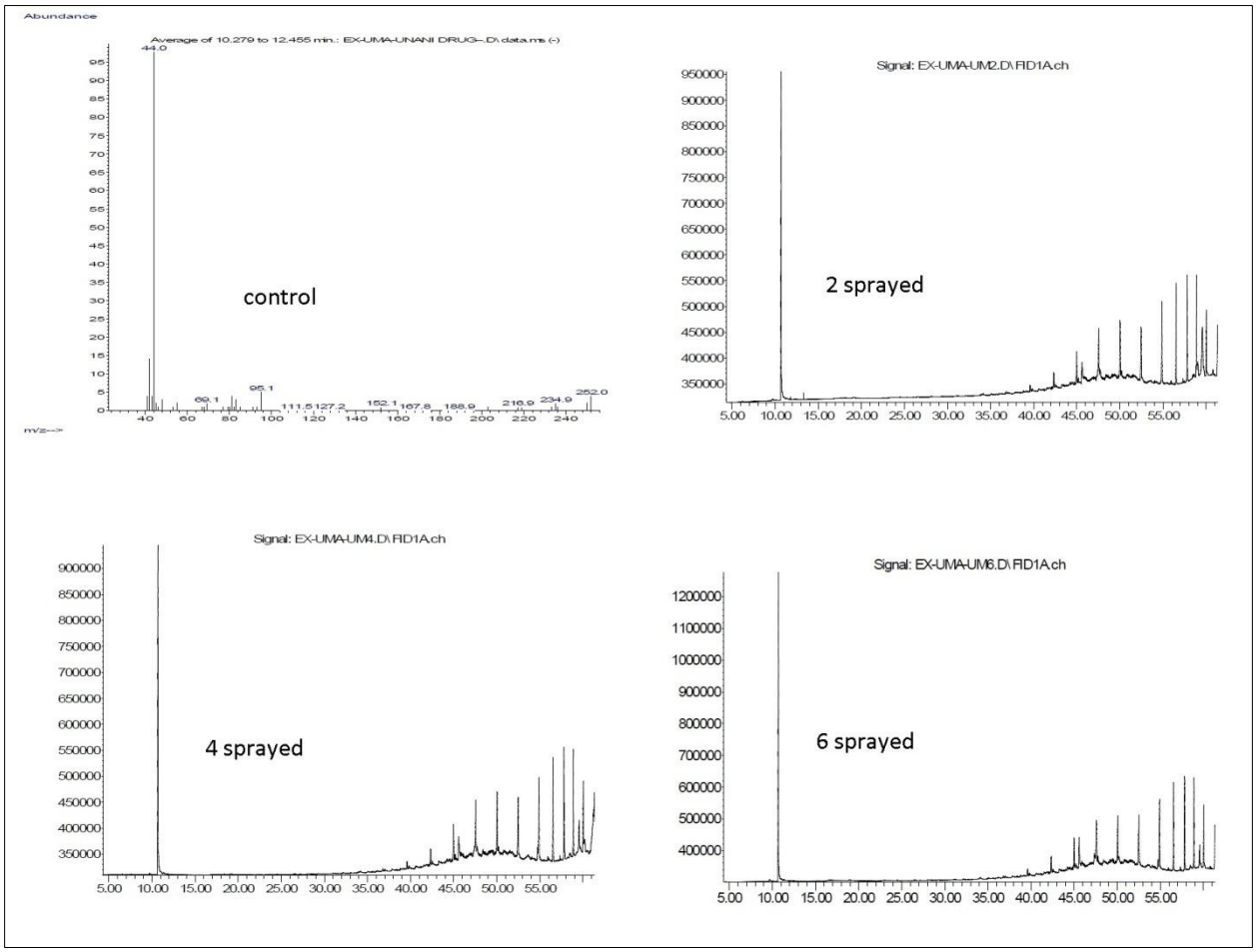

Figure 3 GC-MS of control and sprayed *Marham Kafoori*

Control sample contains Hexadecanoic acid, 1-(hydroxymethyl)-1, 2-ethanediyl ester, Octadecane, 2-methyl, 1-Iodo-2-methylundecane as significant peaks. In2 sprayed sample, there were new peaks of Ethyl iso-allocholate & Ethanol, 2-(octadecyloxy), and an increase in the peak of Eicosane. Additionally, there was no peak of Hexadecanoic acid, 1-(hydroxymethyl)-1, 2-ethanediyl ester and Ergosta-5, 22-dien-3-ol, acetate, (3β, 22E). These differences are responsible

for increased potency than control. On the other hand, 4 sprayed sample has shown an abundant peak of Ethanol, 2-(octadecyloxy), but has a lesser amount of Ethyl iso-allocholate than 2 sprayed sample. There was no peak Hexadecanoic acid, 1-(hydroxymethyl)-1, 2-ethanediyl ester and Ergosta-5, 22-dien-3-ol, acetate, (3β, 22E) as compared to control. There was an increase in Eicosane peak in 4 sprayed sample. these all contributed to the tremendous increased in the potency of the drug as compared to control and 2 sprayed sample. In 6 sprayed sample, the most abundant peak was Bicyclo[2.2.1]heptan-2-one, 1,7,7-trimethyl-, (1S). However, the peak of Ethanol 2-(octadecyloxy) was less abundant than 4 sprayed sample. These are the reasons for the decrease in potency of drugs than control. *(Fig 3) (Table 1)*FTIR

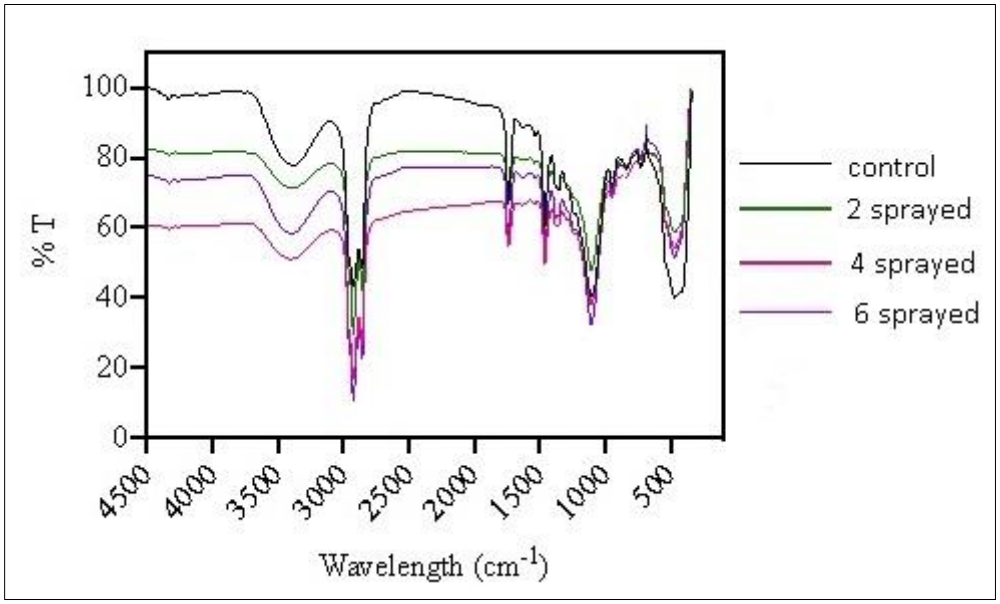

Figure 4 FTIR of control and sprayed *Marham Kafoori*

Table 2 FTIR spectral peak analysis

Peaks (cm^{-1})	***Marham Kafoori***			
	control	2 sprayed	4 sprayed	6 sprayed
640	-	SO_4	SO_4	-
732	= C–H	= C–H	= C–H	= C–H
833	CH_2	-	-	-
840	-	CH_2	CH_2	CH_2
948	C-C	C-C	C-C	C-C
1111	C–O	C–O	C–O	C–O
1242	C–O	-	C–O	C–O
1296	-	CH_2	CH_2	-
1350	CH	-	-	-
1373	-	CH	CH	CH
1465	CH	CH	CH	CH
1535	C=C	C=C	C=C	C=C
1627	H_2O	H_2O	H_2O	H_2O
1735	-	C=O	C=O	-
1743	C=O	-	-	C=O

2845	CH_2	CH_2	CH_2	CH_2
2924	CH_2	CH_2	CH_2	CH_2
3387	H_2O	H_2O	H_2O	H_2O
3394	-	H_2O	H_2O	H_2O
3757	H_2O	-	-	-

In Triple bond region (2700–1950 cm^{-1}), no peaks were observed in any of the samples which may indicate absence of highly complex structure. The region between 1650 and 1450 cm^{-1} provide important information about aromatic rings. Nearly all samples showed either one or more peak in this region. The peaks appear below 500 or above 4000 cm-1 are not considered in this interpretation.

The main difference between these 4 sample is at 640 and 1296 cm^{-1}. Peak at 1296 cm^{-1}due to the CH_2 is missing in the control, it increases upon 2 and 4 sprayed samples, and it again disappears in 6 sprayed sample. This follows the potency behaviour of the sample. The potency of the samples increases upon 2 and 4 sprayings. This suggests breakage of the molecule chain to smaller molecules, hence more potent and active. By continuing the sprayings (x6) the smaller chains again aggregate and form a bigger molecule (less potency). The peaks at 640 cm^{-1}also follows the same pattern. This is mainly due to a head group, such as SO_4. *(Fig 4) (Table 2)*

4.1.2. 1H-NMR

Peaks from the NMR spectrum observed and their origin are summarized along with their integral value in the below *Table 3*. *(Fig 5)*

Table 3 1H-NMR of Marham Kafoori - Peaks and their integral values

Code	**Chemical shift (ppm)**	**Assignment**	**Integral values**			
			Control	2 sprayed	4 sprayed	6 sprayed
A	0.71-1.22	Methyl groups	0.54	0.02	0.76	0.03
B	1.49-2.04	Aliphatic protons	0.06	0.05	0.14	0.01
C	2.27-5.00	Aliphatic protons next to other chemical groups (OH, C=O…)	8.79	0.20	12.70	0.21
D	5.00-5.92	Olefinic protons (alkenes and cycloalkenes)	0.03	< 0.01	0.04	< 0.01
E	6.86-7.94	Aromatic protons	0.01	< 0.01	< 0.01	< 0.01

Comparing the ^{1}H-NMR spectra of the four samples some differences can be observed. Peaks in the region A (0.71-1.22 ppm) are mainly originated by compounds with methyl and saturated chains, typically fatty acids. Active compounds of this kind of drugs are terpenoids and these compounds possess diverse structures, but their proton nuclei are usually concentrated in the region between 1.5 and 5.00 ppm (alkanes, cycloalkanes, alkenes, and protons next to other chemical groups, such as carbonyls).

MIRGA spraying caused an increased activity of the drug, pointing to an augmentation in the concentration of the active components. This is explained by the fact that some non-active components of the sample could be loss or degraded upon mid-IR radiation, causing a net increase in the concentration of active compounds. This is supported by comparing the integral values of regions B and C, where most active compounds are expected to display their signals, a clear difference in the integral values is observed between the control and sprayed samples.

The activity of the drug diminished drastically with six number of sprayings. It is inferred that, an excess of spraying lead to the loss or degradation of part of the active components, which translates into a lower efficacy of the drug.

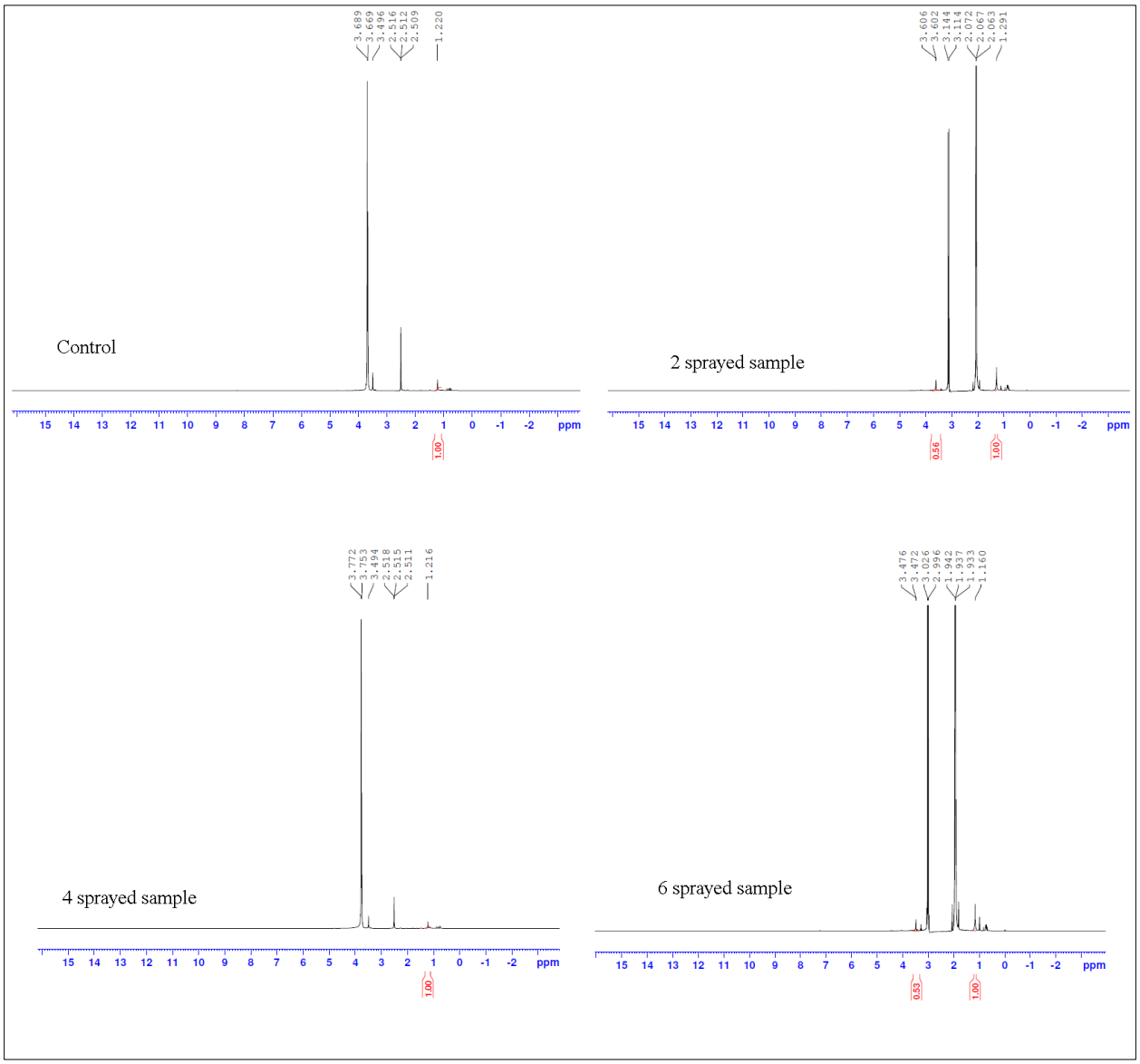

Figure 5 1H-NMR spectra of control and sprayed *Marham Kafoori*

4.2. Invention background

Intermolecular and intramolecular bonding makes up the four visible states of matter: solid, liquid, gas, and plasma. While protons, neutrons, and electrons all have distinctive properties by nature, the differences in their quantities are what distinguish different atoms from one another and determine how these atoms combine to form distinct molecules with individual traits. In the electromagnetic wave (EMW) spectrum *(Fig 6)* mid-IR region is vital and interesting for many applications since that region coincides with the internal vibration of most molecules (CORDIS, European commission). Sixty-six percent of the energy that the sun emits is infrared (Aboud et al., 2019), which is absorbed and radiated by all particles on Earth. The majority of thermal radiation that reaches the Earth's surface is in the mid-IR range. Naturally, at molecular level, interaction of mid-IR wavelength energy elicits rotational and vibrational modes (from about 4500-500 cm^{-1} roughly 2.2 to 20 microns) through a change in dipole movement leading to chemical bond alteration (Girard, 2014).

Our research revealed the following: (A) Although atoms in all objects always remain atoms, their chemical bond parameters are constantly susceptible to change due to cosmic and physical energies (such as heat, pressure, humidity, and EMW). This can result in the bonds breaking (McMakin, 2011; Moss, 2011), stretching/bending (Alvarez et al., 2012; Smith, 1999; Shankar, 2017; Mohan, 2004), and new bond formation (Raven, 2012). These modifications ultimately cause the objects' physicochemical characteristics to change. (B) The dynamic, ongoing, mutual effects of EMW between Earth, celestial bodies, and living things are always changing the physiochemical properties that are intrinsic to Earthly

objects. These properties include enhancement brought about by an ideal energy dosage or decrease/destruction brought about by an excess of energy (explained below). Based on these ideas, the MIRGA was developed to change bond parameters and enhance the inherent qualities of any usable.

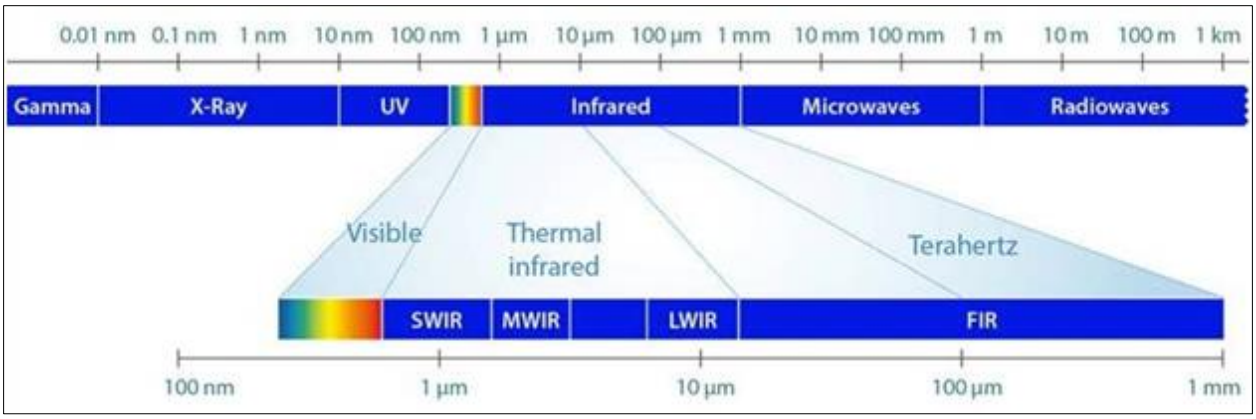

Figure 6 Electromagnetic Spectrum

4.3. MIRGA definition

We define MIRGA as *'a harmless, economical atomizer containing an imbalanced ratio of ions suspended in water, which influence the natural potency of target substances by generating mid-IR while spraying'*.

4.4. Technique of mid-IR generation from MIRGA

We designed MIRGA as to accommodate an imbalanced ratio of ions suspended in water in their fundamental state and can move as free particles. The solution has very little background frequency of detectable disintegration which is less than that of cosmic events whereas even humans have more radioactivity (around 10 microns) (Ashcroft, 2000; Sanders, 2014). We designed *MIRGA to generate energy based on various below given processes like,* (A) spraying leads to ionization (electron getting separated from atom) and the pathway for electron re-absorption are also many, due to these two oscillatory processes energy generated. (B) while spraying, water-based ionic solution gets excited/charged, which in turn leads to oscillation among the imbalanced ions (Verheest, 2000) in their excited state, resulting in the emission of photons (Keping *et al.*, 2004; Fauchais *et al.*, 2014). (C) Though low electromagnetic field exists between charged particles of the MIRGA's ionic solution, during spraying the induced oscillation between these charged particles produces energy (Wendish *et al.*, 2009; Singh, 2009; Prasad, 2017; Pople, 1999). (D) Also, in the natural rainfall process, more energy is required to break water bonds for creating smaller water droplets from the clouds (Barry *et al.*, 1998). Therefore, these droplets should have more stored energy and then travels down at a velocity from a specific distance thus gaining also a kinetic energy. When the rain hits the earth's surface, it forms a very thin film of mid-IR (nearly 6 micron), hence there is a net heat gain (Barry *et al.*, 1998; Eniday, 2019). We simulated this rainfall's energy gaining process also in MIRGA i.e., when imbalanced ions in liquid media are atomized, the ejected smaller droplets should have higher internal energy as well as an acquired kinetic energy and the energy emitted by breaking the surface tension. From trial and experience, we calibrated an ejection pressure to obtain a desired fine mist, and minimized the evaporation rate by altering the pH and density of the solution. Also considering other facts like, the accelerated ions in the sprayed ionic clouds collide among them and generate energy (Krishnakumar, 2019), we incorporated those phenomena in our atomizer and designed in such a way to emit energy in the 2-6 µm mid-IR range.

4.5. Action of MIRGA emitted 2-6 µm mid IR on Marham Kafoori

Depending on number of MIRGA spraying (energy given), a receptor's chemical bond configurations and subsequent physical and chemical characters can be altered to our desire. Similar successful results using MIRGA spray are documented by Umakanthan *et al*., 2022a; Umakanthan *et al*., 2022b; Umakanthan *et al*., 2023c in coffee, tea, cocoa and edible salt. Umakanthan *et al*., 2023d, employed MIRGA and successfully potentiated siddha medicine.

In nature, medicines are biomolecules held up by chemical bonds (Alberts *et al*., 2002), their constant vibration generate infrared radiation (Dakin *et al.*, 2018; de Vries, 1994). When MIRGA is applied over Marham Kafoori, the 2-6 µm mid-IR gets absorbed (Sommer *et al*., 2011), vibrational changes occur (Tsai *et al.*, 2017) leading to chemical bonds parameter changes (Agarwal *et al*., 2014; Mohan, 2004), transformation of chemical compounds and thereby physicochemical modifications **(Yi, 2012;** Datta *et al*., 2014). Thus potentiated and made the Unani medicine favourable. The 2-6 µm

mid-IR is biologically safe and can penetrate intervening media (Pereira *et al.*, 2011; Prasad, 2005). The inorganic compounds used in the generation of MIR are a perspective for biomedical applications (Tishkevich *et al*., 2019; Dukenbayev *et al*., 2019). It is also a new synthesis method for preparation of functional material (2-6 μm mid-IR) (Kozlovskiy *et al*., 2021; El-Shater *et al*., 2022). It is well known that the combination of different compounds, which have excellent electronic properties, leads to new composite materials, which have earned great technological interest in recent years (Kozlovskiy and Zdorovets*, 2021;* Almessiere *et al*., 2022). The MIRGA technology is safe, economical, easy and can be applied over the packaged medicine.

4.6. Toxicological study on MIRGA

Even though, MIRGA generates the safe 2-6 μm mid-IR energy, and moreover spraying is done 0.25–0.50 meter externally right away to packaged consumables, we also wanted to study the MIRGA's toxicity effect by cytotoxicity assay. *In-vitro* Vero, A549 and Human dermal fibroblast cells study proved that MIRGA sprayed mist was non-toxic in any way *(Fig 7)*.

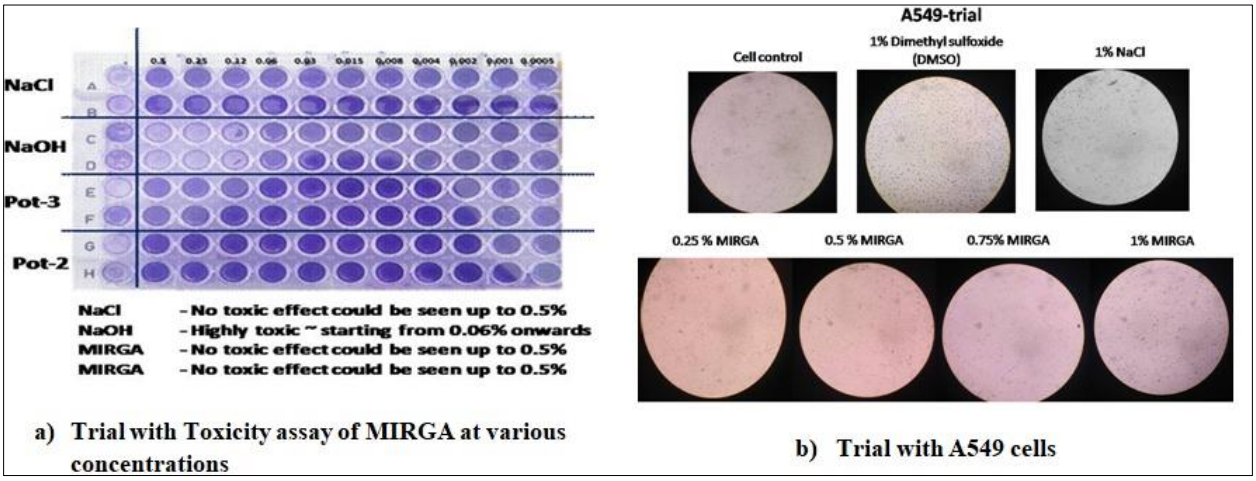

Figure 7 MIRGA's toxicological studies

Field studies also showed that, MIRGA spray is eco-friendly, non-toxic, non-irritant to soft tissues such as cornea, safe to infants even if sprayed directly, needs no skill but easy to handle (like perfume body spray), and highly economical (USD 0.30 per MIRGA unit which emits 300 sprayings).

4.7. MIRGA's past interests

The MIRGA's chemical formulation, therapeutic effect, and results were found to be strikingly similar to those of "The Superior Medicine" found in many ancient medicinal systems, including "Muppu" (Tamil Siddha), "al-Kimiya" (Arabic), "Rasayana" (Indian Ayurveda), "Rasavatan" (Persian), Materia prima, philosopher's stone, tincture (Europe), and Taoist alchemy, hudan, or Jindan (Chinese). Furthermore, MIRGA spraying and hand healing therapy that uses body-generated infrared are comparable. This MIRGA based on water may be the first new pioneering technology to be potentiated. According to the Blue Planet Project, atomizer technology of this kind appears to be utilised by extraterrestrials for therapeutic purposes during visits.

4.8. MIRGA's future scope

We have achieved a range of 30% to 173% potentiation in different usables. Consideration of even the lowest 30% in certain useful items has led to 30% savings in terms of money, resources, and the environment, along with positive health effects. However, there is still a knowledge gap about potentiation from 30% to at least 100% for all consumables. This knowledge can be filled-up by refining MIRGA's ionic solution, concentrations, atomizer pressures, other parameters and even formulating a better solution.

There is currently a wide range of mid-IR emitters available, such as silicon photonic devices (CMOS Emerging Technologies, 2012), non-cascade based lasers, quantum and interband cascade lasers (Jung et al., 2017), chalcogenide fiber-based photonic devices (Sincore et al., 2018), and suspended-core tellurium-based chalcogenide fibre photonic devices (Bo et al., 2018). These emitters are only used in astronomy, the military, medicine, industry, and laboratories;

they are not as affordable as MIRGA. For the ordinary user, they are far too sophisticated in everyday residential applications.

Given MIRGA's broad spectrum of applications, we think it will be well received in a wide range of scientific studies, including those in the domains of biophotonics, pharmaceuticals, health, ecology, and many more. We are currently conducting dynamically ongoing research on MIRGA and its various developed manifestations in human endeavours, notably MIRGA salt, MIRGA vapour, and MIRGA plasma.

5. Conclusion

The four times mid-IR irradiated *Marham Kafoori* was found to be more effective with 20-40% reduced course of treatment, and thus the MIRGA technology demonstrated to potentiate the marketed Unani formulation. We with nearly two decades of experience, are hopeful that further research using mid-IR will demonstrate the possibility of potentiating other unani drugs also, without the need for developing high potency formulations which incur potential risks in terms of efficacy, toxicity, cost and raw material resources.

Author contribution

- *Umakanthan:* Conceptualization, Methodology, Supervision, Validation.
- *Madhu Mathi:* Data curation, Investigation, Visualization, Writing - Original draft preparation.
- *Umadevi*: Project administration, Resources
- *Umakanthan, Madhu Mathi:* Writing- Reviewing and Editing.

Compliance with ethical standard

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Disclosure of Competing interest

In accordance with the journal's policy and our ethical obligation as researchers, we submit that the authors Dr.Umakanthan and Dr.Madhu Mathi are the inventors and patentee of Indian patent for MIRGA *(under-patent no.: 401387)* which is a major material employed in this study.

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