INTERNATIONAL JOURNAL OF PHARMACY AND ANALYTICAL RESEARCH

ISSN:2320-2831

*IJPAR |Vol.6 | Issue 2 | April- June-2017*
*Journal Home page: www.ijpar.com*

*Research article* *Open Access*

# Physico-chemical analysis of kanakalinga karpurathy mezhugu – A Siddha herbo mineral compound

**V. Elango*, J.Jeya venkatesh**

*Department of Siddha Medicine, Tamil University, Thanjavur, Tamil Nadu, South India.*
***Corresponding Author: Dr. V. Elango**
**Email: drelangovantu@gmail.com**

## ABSTRACT

The present study is aimed at the characterization of the physico-chemical traits of the traditional Indian Siddha medicine, *Kanakalinga karpurathy mezhugu*. FT-IR spectroscopy has been used to study the presence of organic substance compound and complexes in the sample. Proton NMR studies help to characterize the structure of the functional groups in the sample. The study highlights the appropriate application of modern scientific methods for developing new insights into metal based siddha drugs.

**Keywords:**Siddha, Herbo-mineral, Physico-chemical analysis, Kanakalinga Karpurathy Mezhugu, FTIR, Proton NMR, Proton shift

## INTRODUCTION

The Siddha system is an Indian system of medicine which is developed and mainly practised in Tamil Nadu and other parts of Southern India since ancient times [1]. The materiamedica of Siddha system encompasses herbal, mineral, animal, herbo mineral drugs. Siddha medicine depends largely on drugs of metallic origin in contrast to Ayurveda and Unani medicine those less in number. The advantages of metal and mineral based medicines in Siddha are smaller or nano in doses, fast in acute and emergency conditions, can be used for wide range of diseases, high Shelf life, nil chemical changes in geo-climatic conditions and less expensive. A special care is taken to administer and follow-up of therapy with suitable adjuvants, diet habits, post therapy care to nullify the unwanted effects.

Generally, the method of preparation of metal based Siddha medicines involves conversion of minerals or metals into the oxide or sulphide form by various herbal treatments followed by repeated high temperature calcination and grinding cycles from *Suddhi* (purification or detoxication) to finished drug. The *mezhugu* thus obtained constitute ultra-small particles and are taken along with vehicles such as milk, honey, butter, ghee etc according to the disease. This makes these drugs easily assimilable, eliminating their harmful effects and enhancing their biocompatibility [2]. The rigidity of the methods of preparation for a

particular mezhugu makes the drug, unique. However very few studies have been carried out to understand the physico-chemical nature of these type of traditional medicines [3, 4]. Though metal based Siddha medicines are time tested drugs, extensive research works should be carried out to explore its effectiveness and to bring all Indian Traditional Systems into the limelight.

There is a general public apprehension regarding the toxicity of these medicines due to western people apathy to the traditional systems of medicine as evidenced by some of the studies [5-8]. For metal and mineral based medicinal preparations, it becomes imperative that these drugs should be characterized with the help of modern instrumental techniques like Infrared Spectroscopy (IR) and proton NMR. Based on these, the specifications of metal based drugs can be well standardized on a scientific basis. The present study investigated the physico-chemical properties of the traditional Indian Siddha medicine, *Kanakalingakarpurathymezhugu*, which is widely used for treating anaemic conditions, scrofula, headache and rheumatic diseases. It is also used for the treatment of piles, uterine disorders, orchitis, gastric ulcer, syphilis and tuberculosis etc.

## MATERIALS AND METHODS

### Preparation of Kanakalingakarpurathymezhugu

The Siddha medicine *Kanakalingakarpurathymezhugu* was prepared in our laboratory under the experts from Siddha medicine, reference obtained from texts *AnubogaVaidyaNavaneetham Part IV*, *Siddha Maruthuvarin Diary Kurippu* and few traditional methods followed in Kanniyakumari district.

### Preparation of sample

About 20 grams of the *Kanakalingakarpurathymezhugu* sample powdered were soaked in 100 ml methanol individually. It was left for 24 hours so that alkaloids, flavonoids and other constituents if present will get dissolved. The methanol extract was filtered using Whatman No.1 filter paper and the residue was removed. It was again filtered through sodium sulphate in order to remove the traces of moisture. *Kanakalingakarpurathymezhugu* was studied in two phases. The first phase included the study of compound drug before purification and the second phase included the purified finished product.

The studies of the purification methods and effectiveness of the drug and its mechanism of action will be discussed correlating the phytochemical and chemical constituents of the herbal and herbo-mineral drugs and the alterations in the biochemical parameters pertaining to the hormone deficiency disorder in the treatment of the drug.

## METHODS

### I R – Spectral Studies

IR spectral studies are carried out with a view to knowing the presence of organic compounds as impurities in the *Kanakalingakarpurathimezhugu*. The FTIR spectrum of *Kanakalingakarpurathimezhugu* by the major stretching vibrations of different functional groups in organic compounds in the spectra has very low intensity. This shows that *Kanakalingakarpurathimezhugu* is almost free from organic compounds. The presence of low organic matter is ample proof for proper cleanliness during the preparation of these medicines and confirms the absence of any external organic contamination. During the burning process in purification and grinding process to finish the drug involved in the preparation of *Kanakalingakarpurathimezhugu*, the organic groups might have changed into gaseous oxidized compounds and might have escaped. Multitude of absorption signals in the far IR region suggests the presence of metal-oxygen and metal-sulphur linkages.

### $^{1}$H &$^{13}$C NMR spectral studies

*Proton nuclear magnetic resonance* (proton NMR, hydrogen-1 NMR, or $^{1}$H NMR) is the application of nuclear magnetic resonance in NMR spectroscopy with respect to hydrogen-1 nuclei within the molecules of a substance, in order to determine the structure of its molecules. [1] In samples where natural hydrogen (H) is used, practically all the hydrogen consists of the isotope $^{1}$H (hydrogen-1; i.e. having a proton for a nucleus). A full $^{1}$H atom is called protium. *Carbon-13 nuclear magnetic resonance* (most commonly known as carbon-13 NMR or $^{13}$C NMR or

sometimes simply referred to as carbon NMR) is the application of nuclear magnetic resonance (NMR) spectroscopy to carbon. It is analogous to proton NMR (1HNMR) and allows the identification of carbon atoms in an organic molecule just as proton NMR identifies hydrogen atoms. As such $^{13}C$ NMR is an important tool in chemical structure elucidation in organic chemistry. $^{13}C$ NMR detects only the 13C isotope of carbon, whose natural abundance is only 1.1%, because the main carbon isotope, 12 C , is not detectable by NMR since it has zero net spin. Analyses were performed on a Bruker NMR (400 $MH_Z$, MeoD) All (1,3,5-tris [trifluro methyl] benzene). A standard TMS was used to the NMR spectrum of *Kanakalingakarpurathymezhugu.*

## RESULTS AND DISCUSSION

The studies on *Kanakalingakarpurathymezhugu* using I R – Spectral Studies and $^{1}H$ &$^{13}C$ NMR spectral studies show few points about the purity and characteristic of the herbomineral drug.

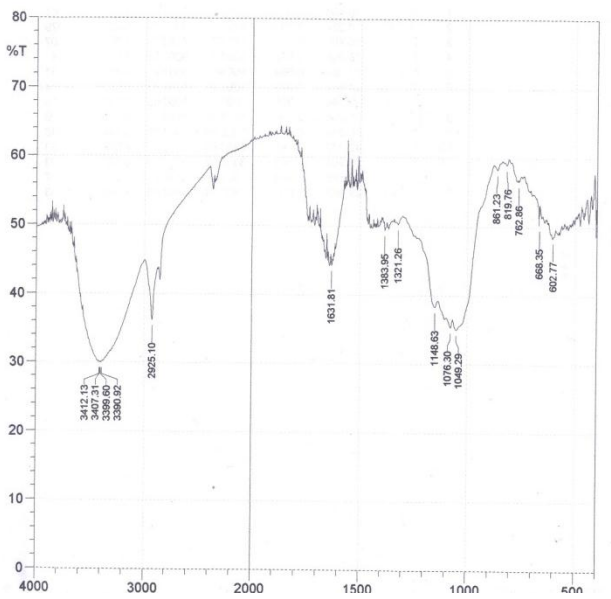
## RESULTS

### I R – SPECTRAL STUDIES

IR spectral studies are carried out with a view to knowing the presence of organic compounds as impurities in the *Kanakalingakarpurathimezhugu*. Shimadgu IR prestige 21 (FT-IR) was used to record the spectrum for the drug under investigation before and after the purification.

### Before Purification

FT-IR spectrum of K*anakalingakarpurathymezhugu* before purification is shown in the fig 1.



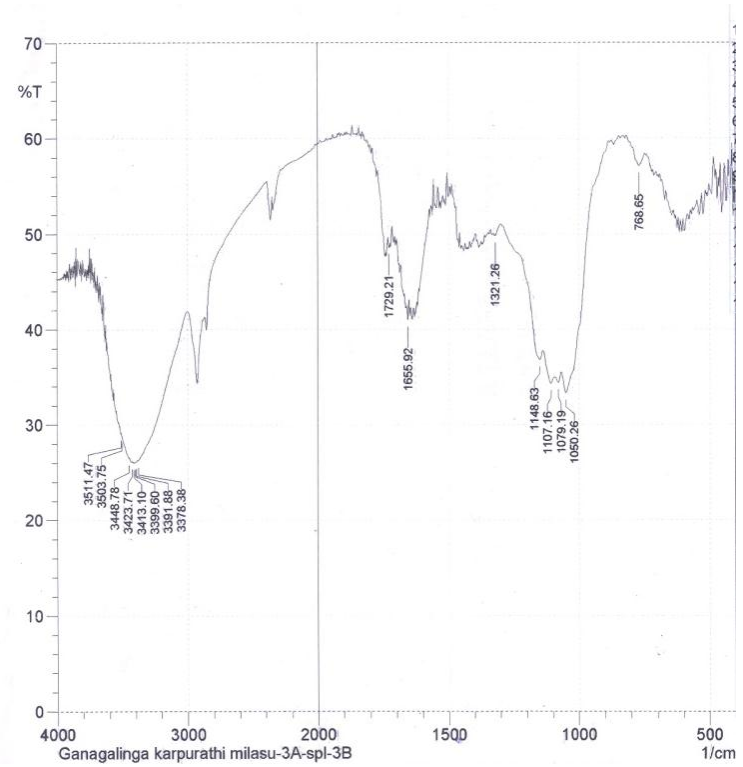



**Figure 1**

## IR Spectrum of *Kanakalingakarpurathymezhugu* (before purification)

The IR data and the corresponding assignments of the functional groups present in the drug before purification are presented in the table 1.

**Table 1**

| Frequency, $cm^{-1}$ | Functional group |
|---|---|
| 3412-3390 | -OH of polymeric alcohol (or) phenol with intermolecular rydnogen bonding |
| 2925 | C-H stretching (arornatic) |
| 1631 | C=0 present in the conjugated system |
| 11-48 | O-H stretching (secondary alcohol) |
| 1078-1049 | C-O stretching and O-H bending coupled |

## IR data of the *Kanakalingakarpurathymezhugu* and the corresponding assignments (before purification)

A strong and broad absorption band at the range 3448 – 3378 $cm^{-1}$ is due to the O-H stretching. Further, this band indicates that the O-H group may be present in the alcohol of polymeric in nature. The bands at 1148 and 1078-1049 $cm^{-1}$ are due to the stretching and banding vibration of C-O and O-H bonds. These absorptions support the presence of alcohol. The strong band at 1631 $cm^{-1}$ shows the presence of carbonyl group which may be in the conjugated system. The place and intensity data of this spectrum is presented in the table 2.

**Table 2**

| | Peak | Intensity | Corr.Intr | Base(H) | Base(L) | Area | Corr.Are |
|---|---|---|---|---|---|---|---|
| 1 | 602.77 | 48.458 | 0.07 | 603.73 | 599.87 | 1.21 | 0.001 |
| 2 | 668.35 | 50.271 | 3.105 | 679.92 | 666.42 | 3.723 | 0.079 |
| 3 | 762.86 | 56.588 | 0.26 | 766.72 | 759.97 | 1.662 | 0.007 |
| 4 | 819.23 | 58.265 | 0.694 | 869.91 | 850.62 | 4.474 | 0.051 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| 5 | 861.23 | 34.997 | 0.992 | 1066.55 | 1034.83 | 14.324 | 0.214 |
| 6 | 1049.29 | 34.997 | 0.992 | 1066.65 | 1034.83 | 14.324 | 0.214 |
| 7 | 1076.3 | 35.348 | 1.256 | 1096.55 | 1066.65 | 13.249 | 0.215 |
| 8 | 1148.63 | 38.258 | 2.297 | 1226.75 | 1135.13 | 33.489 | 0.396 |
| 9 | 1321.26 | 50.238 | 0.591 | 1328.98 | 1311.62 | 5.144 | 0.046 |
| 10. | 1383.95 | 49.376 | 1.281 | 1386.84 | 1373.34 | 4.075 | 0.083 |
| 11. | 1631.81 | 44.268 | 1.326 | 1634.7 | 1626.98 | 2.699 | 0.061 |
| 12 | 2925.1 | 36.142 | 7.98 | 2988.75 | 2876.88 | 43.437 | 3.762 |
| 13 | 3390.92 | 29.969 | 0.059 | 3391.88 | 3385.13 | 3.522 | 0.003 |
| 14 | 3399.6 | 29.896 | 0.133 | 3402.49 | 3396.7 | 3.03 | 0.007 |
| 15 | 3407.31 | 29.983 | 0.069 | 3409.24 | 3403.45 | 3.024 | 0.003 |
| 16 | 3412.13 | 29.973 | 0.117 | 3420.81 | 3409.24 | 6.039 | 0.01 |

## Peak and intensity of FT-IR data of spectrum-1 (before purification)

## After Purification

FT-IR spectrum of the drug after purification is presented in the fig. 2.

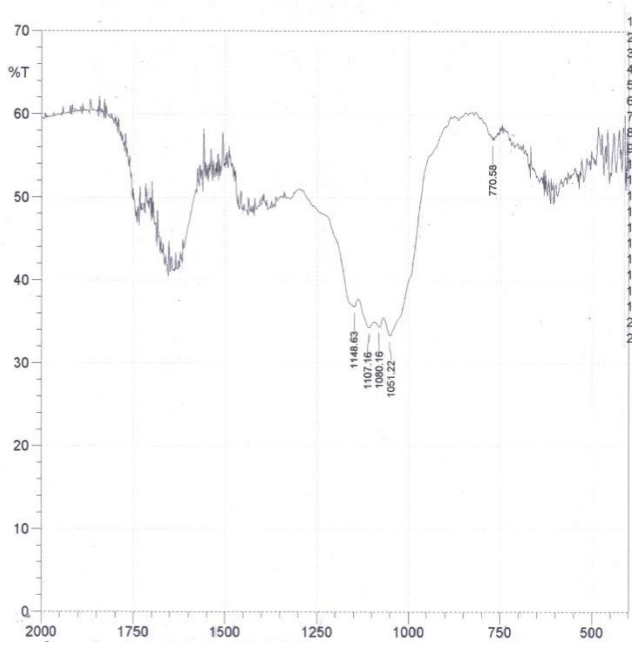



**Figure 2**

## IR spectrum of *Kanakalingakarpurathymezhugu* (after purification)

The IR spectral data and the corresponding functions of the drug are given in the table 3.

**Table 3**

| Frequency, $cm^{-1}$ | Functional groups |
|---|---|
| 3511-3373 | O-H stretching of tree water molecule which coordinates with metal in |
| 1148 – 1061 | O-H plane bending vibration |

## FT-IR spectral data of *Kanakalingakarupurathymezhugu* and corresponding assignments (after purification)

Strong and broad bands at the range 3511-3373 $cm^{-1}$ reveal that the drug may contain tree $H_2O$ molecule which may coordinate with metal ion in the *Kanakalingakarpurathymezhugu*, further the bands at the range 1148-1061 $cm^{-1}$ are due to the banding vibration of O-H bonding. FT-IR peak and intensity data of this spectrum is shown in the table 4.

**Table 4**

| S.No | Peak | Intensity | Corr.inte | Base(H) | Base(L) | Area | Corr.Are |
|---|---|---|---|---|---|---|---|
| 1 | 770.58 | 56.994 | 0.461 | 774.43 | 168.65 | 1.402 | 0.008 |
| 2 | 1051.22 | 33.332 | 4.715 | 1067.62 | 941.28 | 49.261 | 4.506 |
| 3 | 1080.16 | 34.403 | 0.829 | 1090.76 | 1067.62 | 10.611 | 0.13 |
| 4 | 1107.16 | 34.332 | 0.118 | 1137.06 | 1106.19 | 13.746 | 0.059 |
| 5 | 1148.63 | 36.794 | 0.651 | 1158.27 | 1137.06 | 9.132 | 0.087 |
| 6 | 3246.25 | 29.867 | 0.122 | 3248.18 | 3243.36 | 2.523 | 0.005 |
| 7 | 3270.36 | 28.942 | 0.082 | 3272.29 | 3248.18 | 12.815 | 0.009 |
| 8 | 3280.01 | 28.725 | 0.035 | 3280.97 | 3272.29 | 4.689 | 0.003 |
| 9 | 3306.05 | 28.008 | 0.041 | 3307.01 | 3281.94 | 13.733 | 0.021 |
| 10 | 3324.37 | 27.529 | 0.057 | 3326.3 | 3307.98 | 10.195 | 0.004 |
| 11 | 3337.87 | 27.06 | 0.077 | 3338.84 | 3326.3 | 7.064 | 0.003 |
| 12 | 3350.41 | 26.807 | 0.034 | 3351.37 | 3343.66 | 4.397 | 0.002 |
| 13 | 3359.09 | 26.619 | 0.032 | 3360.05 | 3352.34 | 4.422 | 0.001 |
| 14 | 3369.7 | 26.382 | 0.03 | 3370.66 | 3361.02 | 5.562 | 0.003 |
| 15 | 3375.49 | 26.27 | 0.026 | 3376.45 | 3371.63 | 2.795 | 0.001 |
| 16 | 3389.95 | 26.122 | 0.072 | 3390.92 | 3383.2 | 4.488 | 0.006 |
| 17 | 3398.63 | 26.014 | 0.117 | 3402.49 | 3395.74 | 3.943 | 0.008 |
| 18 | 3413.1 | 26.009 | 0.077 | 3418.88 | 3410.2 | 5.072 | 0.009 |
| 19 | 3421.78 | 26.098 | 0.058 | 3425.64 | 3419.85 | 3.374 | 0.003 |
| 20 | 3447.82 | 26.388 | 0.34 | 3460.35 | 3445.59 | 8.312 | 0.04 |
| 21 | 3503.75 | 28.695 | 0.349 | 3507.61 | 3501.83 | 3.125 | 0.02 |

## IR spectral peak and intensity Fig. 2 (After Purification)

FT-IR data of *Kanakalingakarpurathymezhugu* after purification indicate the absence of organic compounds in this drug.

## $^{1}H$ &$^{13}C$ NMR spectral studies

The purity of the drug is further proved by $^{1}H$ &$^{13}C$ NMR studies. These spectra of the drug were recorded on Bruker 400 MHz using methanol-d as solvent and Tetramethylsilance (TMS) as standard.

### Before Purification

$^{1}H$ NMR spectrum of the drug is shown in the fig 3.

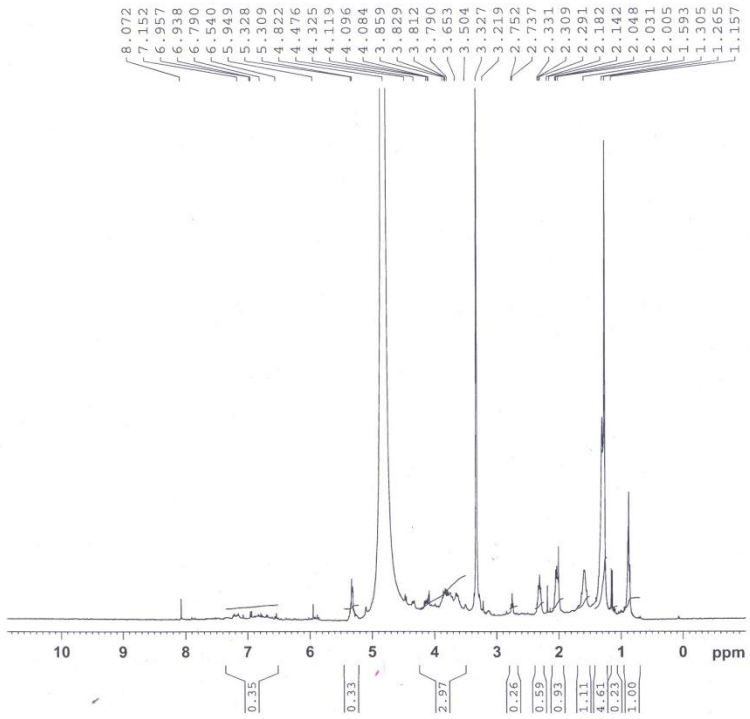



**Figure 3**

## [1]H NMR Spectrum of Kanakalingakarpurathymezhugu (before purification)

[1]H NMR data and the corresponding assignments are given in the table 5.

**Table 5**

| Chemical Shift, S, ppm | Assignment |
|---|---|
| 8.01-5.94 | C-H(aromatic) |
| 5.34(s) | $-CH_2-Z$ (Z= electronic with drawing group) |
| 4.21-3.21(m) | C-OH (alcohol/water) |
| 1.59(s) | $-CH_3$ |
| 1.39-1.26(d) | $-CH_2-CH-$ |
| s=Singlet, d = doublet, m=multiplet | |

## [1]HNMR chemical shift value and the corresponding assignment of *Kangalingakarpurathymezhugu* (before purification)

The chemical shift values at the range is 8.01 to 5.94 ppm are due to the aromatic protons which proved the presence of aromatic organic compounds. The multipled at S 4.21-3.21 ppm proves the presence of O-H of water and alcohol. A songlet at S 5.34 ppm reveals the presence of $-CH_2-$ group such as $-No_2$, $-Oc_2H_5$, -OH etc. similarly there is a singlet at S 1.59 which shows the presence of –CH3 group in the organic compound. The signal appeared at S1.361.26 ppm as doublet is for the –CH- which is attached with $-CH_2-$ group.

$^{13}$C NMR spectra of this drug is presented in the fig 4.

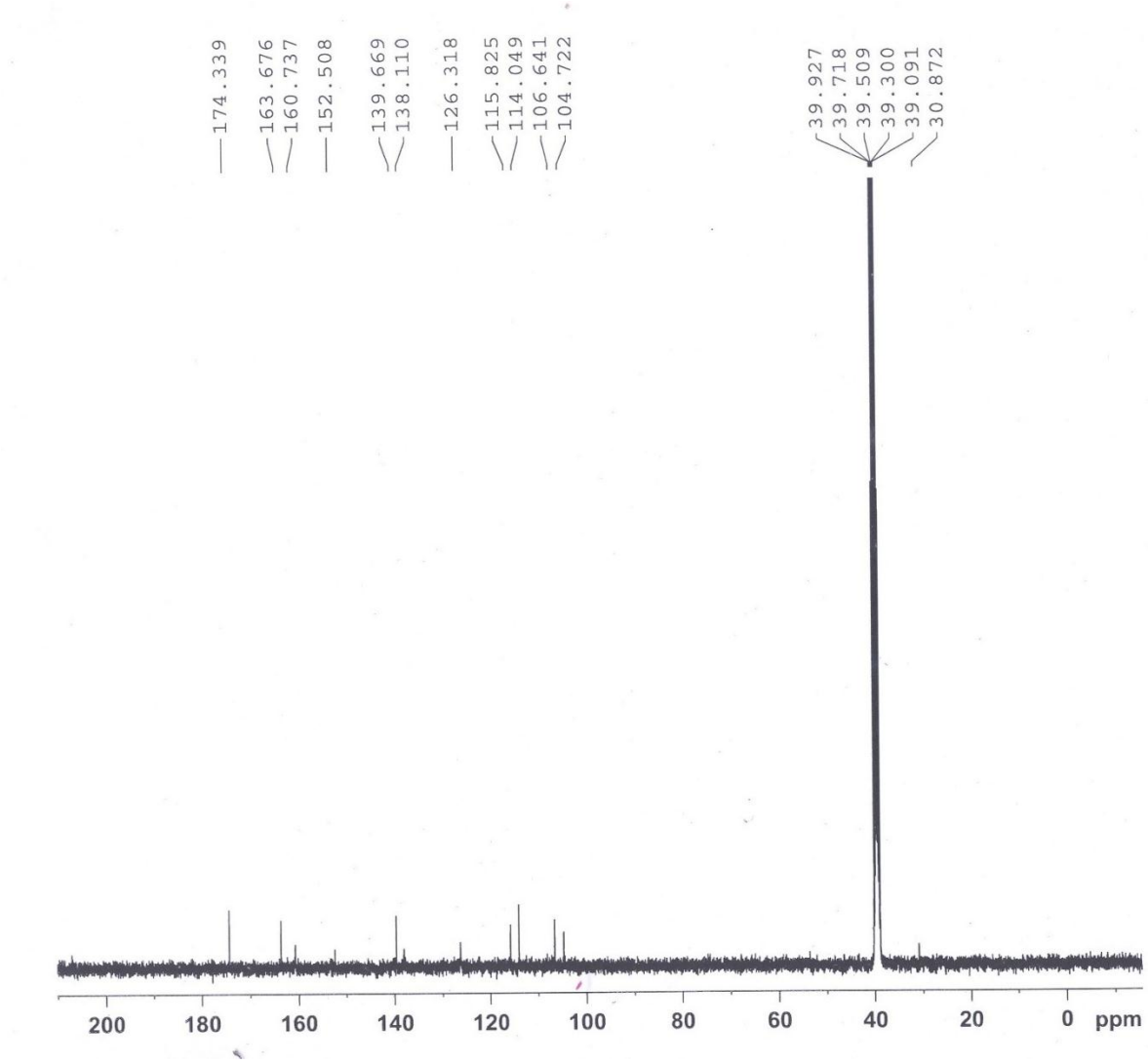



**Figure 4**

## $^{13}$C NMR spectrum of *Kanakalingakarpurathymezhugu* (before purification)

The chemical shift values and the corresponding assignments are presented in the table 6.

**Table 6**

| Chemical Shift, S, ppm | Assignment |
|---|---|
| 174 | C of C=0 group |
| 163,160,152,139,138,126,115,114,106 & 104 | Aromatic carbons |
| 33,30,26,23 & 14 | aliphatic carbons |

## $^{13}$C NMR spectral data Kanakalingakarpurathymezhugu and the corresponding assignments (before purification)

The chemical shift values account for the presence of aromatic and alphabetic compounds with carbonyl group.

## After purification

$^{1}$H NMR spectrum of the drug is shown in the fig 5.

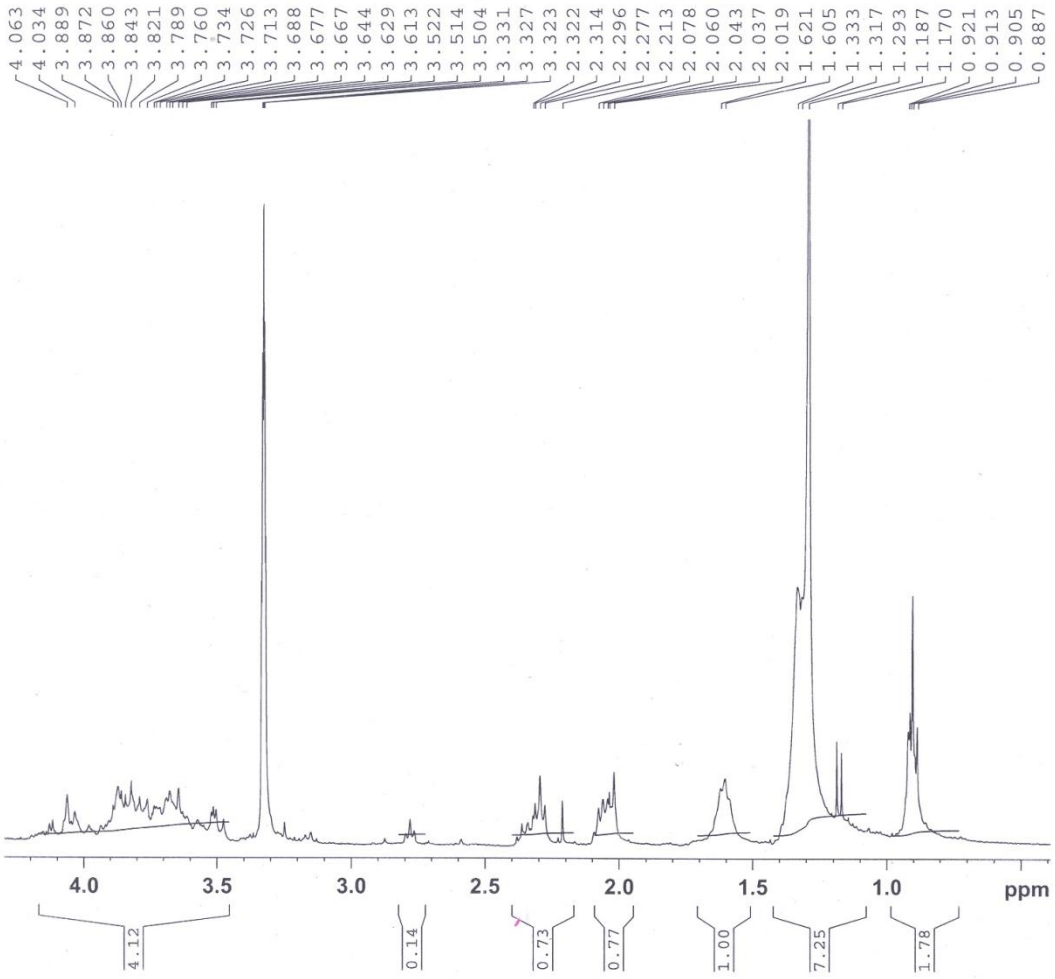



**Figure 5**

## $^{1}$H NMR spectrum of *Kanakalingakarpurathymezhugu* (after purification)

The chemical shift values and the corresponding assignments are presented in the table 7.

**Table 7**

| Chemical Shift, S, ppm | Assignment |
|---|---|
| 3.88-3.32(m) | O-H of water and C-OH of alcohol |
| 2.32-2.27(q) | $-CH_2 - CH_3$ |
| 2.07-2.01(m) | $(CH_2)_n$ (n=1,2,...) |
| 1.62-1.60(d) | $-CH_2-{}^{1}CH$ |
| 1.33-1.29(t) | $-CH_2-CH_3$ |

### [1]HNMR spectral data of *Kanakalingakarpurathymezhugu* and the corresponding assignments (after purification)

The chemical shift values at the range S4.06-3.32 ppm are due to O-H and C-OH protons water and alcohol. The chemical shift occur at S2.32-2.27(q) 2.07-2.01(m), 1.62-1.60(d) and 1.33-1.29(t) ppm are due to the presence of $–CH_3$, $-CH_3$, $-CH_2-$ and –CH- groups. The very low intensity of the peaks indicate the presence of the brace amount of organic compounds in the drug.

[13]CNMR spectrum of *Kanakalingakarpuarathymezhugu* is shown in the fig. 6.

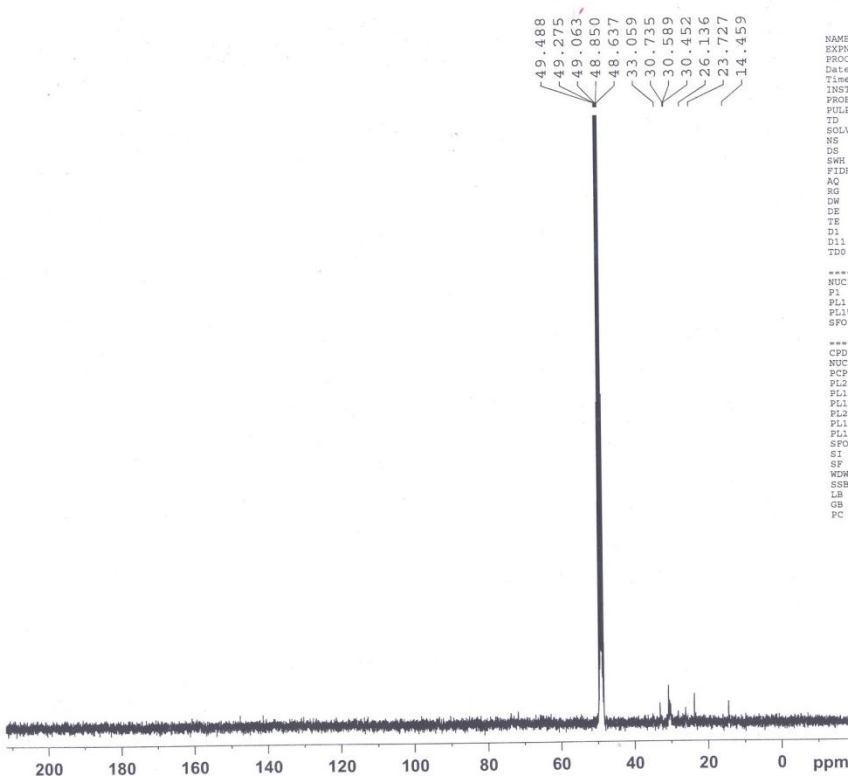



**Figure 6**

### [13]CNMR spectrum of *Kanakalingakarpurathymezhugu* (after purification)

The chemical shift values and the corresponding assignments are shown in the table 8. The removal of organic compounds from the drug is known from the spectral data.

**Table 8**

| Chemical shift S, ppm | Assignment |
|---|---|
| 33,30,26,23 & 14 | Alkyl carbons |

### [13]CNMR spectral data and the corresponding assignments for the Kanakalingakarpurathymezhugu (after purification)

## DISCUSSION

From the spectral (IR, ‘H &[13]C NMR) data after the purification indicates the presence of negligible quantity or almost nil organic compound in the purified drug which is the proof for the purity of the drug. During the purification of the drug *Kankalingakarpurathymezhugu* by burning process

the organic compounds might have changed into oxidized compound and have gone to gaseous state.

Analyses were performed on a Bruker NMR (400 $MH_Z$, MeoD) All (1,3,5-tris [trifluro methyl] benzene). A standard TMS was used to the NMR spectrum of *Kanakalingakarpuratymezhugu* before purification in presented in the figure 1, doubling or tripling the sample concentration increases the signal strength proportionally very low. This shows that *Kanakalingakarpurathymezhugu* is almost free from organic compounds. The presence of low organic matter is ample proof for proper cleanliness during the preparation of these medicines and confirms the absence of any external organic contamination. During the burning process involved in the preparation of *Kanakalingakarpurathymezhugu*, the organic groups might have changed into gaseous oxidized compounds and might have escaped.

## CONCLUSION

The present study evaluated the physico-chemical properties of the traditional Indian medicine *Kanakalingakarpurathymezhugu*. The results of XRD, ICP, FTIR and SEM studies can be used as excellent physico-chemical fingerprints for the validation of the medicine. The near-nano size of *Kanakalingakarpurathymezhugu* may enable better bio-absorption. Drugs in Indian system of medicine are hesitated by other counties due to the poor standardization and lack of quality. This study is an earnest attempt of bio active principles present in the drug, in relation with their actions at making appropriate scientific validation of metal based ancient Siddha medicine using authentic scientific techniques.

## REFERENCES


[1]. Scott Hajicek-Dobberstein. Soma siddhas and alchemical enlightenment: psychedelic mushrooms in Buddhist tradition. J Ethnopharmacol 48, 1995, 99-118.

[2]. Suoboda RE. Prakriti: your ayurvedic constitution. MotilalBanarsidassPubl, India 1998.

[3]. Kumar A, Nair AGC, Reddy AVR, Garg AN. Bhasmas: unique ayurvedic metallic-herbal preparations, chemical characterization. Biol Trace Elem Res 109, 2006, 231-254.

[4]. Wadekar MP, Rode CV, Bendale YN, Patil KR, Gaikwad AB, Prabhune A. Effect of calcinations cycles on the preparation of tin oxide based traditional drug: Studies on its formation and characterization. J Pharmaceu Biomed Anal 41, 2006, 1473-1478.

[5]. Saper RB, Kales SN, Paquim J, Burns MJ, Eisenberg DM, Davis RB, Phillips RS. Heavy Metal Content of Ayurvedic Herbal Medicine Products. J AM MED ASSOC 292, 2004, 2868-2873.

[6]. Grippo AA, Hamilton B, Hannigan R, Gurley BJ. Metal content of ephedra-containing dietary supplements and select botanicals. Am J Health Syst Pharm 63, 2006, 635-644.

[7]. Hardy AD, Sutherland HH, Vaishnav R, Worthing MA. A report on the composition of mercurials used in traditional medicines in Oman. J Ethnopharmacol 49, 1995, 17-22.

[8]. Ernst E. Heavy metals in traditional Indian remedies. Eur J ClinPharmacol 2002; 57: 891-896.

[9]. Dr.Ka.Su.Utthamarayan, Siddha AruvaiMaruthuvam, Dept. of Indian Medicine and Homeopathy, Chennai 106, 1999.

[10]. DrKa Na KuppusamyMudaliyar, Siddha Maruthuvam, Dept. of Indian Medicine and Homeopathy, Chennai 106, 2007

[11]. T V SambasivamPillai Tamil-English Dictionary,The Research Institute of Siddhar's Science, Mount Road, Madras 1931

[12]. Dr Ma Shanmugavelu, NoiNaadalNoiMuthalNaadalThirattu, Dept. of Indian Medicine and Homeopathy, Chennai 106, 2003.

[13]. KanthasamyMudaliar, Atmaratchamirthamennumvaidya Sara Sangiragam, Jeyam Print, Madurai, 2003

[14]. Pu. Su KuppusamyMudaliya, AnubogaVaidyaBirammaRagasiyamr, 2002

[15]. PatharthaGunaChinthamani, Dept. of Indian Medicine and Homeopathy, Chennai 106, 2007.

[16]. MunusamyMudaliyar, MooligaiMarmam, Arangasamynaicker sons, Chennai 1939.

[17]. Dr. M. Sourirajan, Sarabendrar Siddha maruthuvaSudar, SaraswathyMahalNoolagam, Thanjavur 2007.

[18]. Dr.Ka.Su.Utthamarayan, ThotraKiramAraichiyum Siddha maruthuvavaralarum, Dept. of Indian Medicine and Homeopathy, Chennai 106, 2010.
[19]. Pe.MuRajagopalan, Siddha MaruthuvaPadapPayirchinool, Siddha gurugulam, 1997.
[20]. Dr R Thiagarajan, GunapadamThadhuSeevaVaguppu, Dept. of Indian Medicine and Homeopathy, Chennai 106, 1968.
[21]. DrKaSaMurugesaMudaliyar, GunapadamMooligaiVaguppu, Dept. of Indian Medicine and Homeopathy, Chennai 106, 2003.
[22]. GunapadamMooligaivaguppu
[23]. Hakkeem Pa Muhammadu Abdullah Sayabu. Part IV, Anupogavaidyanavaneetham, TamaraiNoolagam, Chennai.