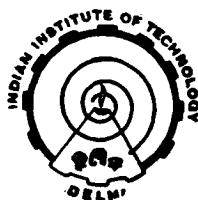


# **STUDIES ON LIQUID AND SUPERCRITICAL FLUID EXTRACTION OF NATURAL PLANT PRODUCTS**

*By*  
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*A THESIS SUBMITTED TO THE  
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FOR THE AWARD OF THE DEGREE OF  
DOCTOR OF PHILOSOPHY*



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## CERTIFICATE

This is to certify that the thesis entitled, "STUDIES ON LIQUID AND SUPERCRITICAL FLUID EXTRACTION OF NATURAL PLANT PRODUCTS" being submitted by Mr. Satya Narayan Naik to the Indian Institute of Technology, Delhi for the award of Doctor of Philosophy is a record of bonafide research work carried out by him under my guidance and supervision in conformity with the rules and regulations, of Indian Institute of Technology, Delhi.

The results contained in this thesis have not been submitted, in part or in full, to any other university or Institute for the award of any degree or diploma.



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(S.N. NAIK)

## NOTATIONS

SCF	=	Supercritical fluid
SC-CO <sub>2</sub>	=	Supercritical carbon dioxide
TLC	=	Thin Layer Chromatography
G.C.	=	Gas chromatography
H P	=	High pressure
min	=	minute
max	=	maximum
hrs	=	hours
n m	=	nanometer
M P	=	Melting point
B P	=	Boiling point
D	=	Diffusion coefficient
r	=	radius
r <sub>o</sub>	=	average radius of spherical particle/half thickness of flake particle/radius of cylindrical particle.
E <sub>1cm</sub>	=	molar absorptivity of unknown compound
Y	=	percentage yield
Y <sub>h</sub>	=	highest yield
Y <sub>∞</sub>	=	percentage yield after infinite extraction time
U	=	fraction of essential oil extracted at any time.
KJ	=	Kilo Joules
mcg	=	micro gram
Sp.gr.	=	Specific gravity
mm	=	millimeter
m	=	meter
i.d.	=	internal diameter
o.d.	=	outer diameter

## ABSTRACT

In the recent years, there has been great interest in liquefied and supercritical gas extraction in chemical industries. In view of this, the status of conventional and liquid/SCF extraction processes have been critically reviewed with special reference to essential oils, food flavours and pharmaceutical substances. Accordingly varieties of plant materials of Indian origins were selected for extraction/fractionation of essential oils and pharmacologically active compounds in liquid/SCF. The selected plant materials were classified in three categories, viz

**Spices:** cardamom, celery, cumin, cumin black, chilli, clove, ginger, fennel, mace, parsley and turmeric.

**Perfumery Plants:** rosemary, sage, sandalwood and vetiver.

**Medicinal:** Kalmegh, malkanguni, periwinkel and Jatamansi.

Two apparatus have been designed to study above mentioned plant materials in liquid/SCF extraction process. 1. High Pressure soxhlet apparatus for liquefied gas extraction. 2. Laboratory model liquid/supercritical fluid extraction apparatus for process optimization and fractionation.

These two apparatuses were fabricated by taking all safety measures into consideration in accordance with safety rules and regulations.

The percentage yields with different extraction time for various plant materials were determined in liquid CO<sub>2</sub> extraction and a mathematical equation for calculating the percentage yields have been suggested on the basis of experimental results. The spherical, cylindrical and flake unsteady state mathematical models were used to obtain mass transfer coefficient parameter  $(D/r_0^2)$  in liquid CO<sub>2</sub> extraction. The determined values of  $(D/r_0^2)$  were used to calculate the percentage yields and the computed values of percentage yields were correlated with experimental data.

The extracts obtained in liquid/SCF extraction and conventional processes were analysed and compared by TLC and GC analysis by adopting standard procedure as reported in the literatures. The structure of the crystalline substance obtained from parsley seed was determined as seseline by mass, IR and NMR studies. The extracts of medicinal plants were also subjected to TLC, IR and UV analysis for identifying active components.

A combined process (solvent extraction and ammonization) has been suggested for detoxification of groundnut and cotton seed meals to reduce aflatoxins with in permissible level.

From the above studies, the following points were emerged in favour of liquid/supercritical fluid extraction:

- (i) The yields obtained by liquid  $\text{CO}_2$  extraction is higher than those obtained by steam distillation.
- (ii) The time and energy requirement in liquid  $\text{CO}_2$  extraction process is less than that require in steam distillation process.
- (iii) The analysis of essential oils obtained by liquid  $\text{CO}_2$  extraction process shows less monoterpene hydrocarbons.
- (iv) The active constituents of medicinal plants like andrographolide, malkanaguni, etc. were sucessfully extracted in liquid and  $\text{SC-CO}_2$ . But for alkaloids separation the liquid and  $\text{SC-NH}_3$  have been found suitable.
- (v) Subcritical state is favourable particulary to essential oil extraction.

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