EXTRACTION OF PHYTOCHEMICALS FROM VANILLA AND NEEM USING PRESSURIZED FLUIDS

by

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Submitted

in fulfillment of the requirements of the degree of

Doctor of Philosophy

to the

Indian Institute of Technology, Delhi

February 2010

tochemical ; Vanilla ; Neem.

Dedicated to .....

My Beloved Grandparents

CERTIFICATE

This is to certify that the thesis entitled "**Extraction of Phytochemicals from Vanilla and Neem Using Pressurized Fluids**" being submitted by ***Mr. Jadeja Girirajsinh Chandrasinh*** to the Indian Institute of Technology, Delhi for the award of the degree of **Doctor of Philosophy** is a record of bona fide research work carried out by him under our guidance and supervision in conformity with the rules and regulations of Indian Institute of Technology, Delhi.

The results presented 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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ACKNOWLEDGEMENTS

If the only prayer you say in your life is "Thank You", it would be enough....

- Meister Eckhart

Apart from God's grace, I would like to grab this opportunity to express my token of thanks to all those who directly or indirectly helped, supported, inspired and motivated me throughout this research work.

First and foremost, I owe my deepest gratitude to my supervisors Prof. S. N. Naik and Prof. R. C. Maheshwari. Their excellent guidance, indelible inspiration, constant encouragement, positive feed-back and supporting attitude during each and every step of the present research work were important for the completion of my thesis.

I would like to give my heartfelt thanks to Prof. A. K. Gupta, one of the members of my research committee, for the meticulous guidance, the constructive criticism and the thought-provoking nuggets I received from him during our discussions. The moral support from him, when I needed it the most, can seldom be expressed in words.

I would also like to thank the other members of my research committee, Prof. Santosh Satya and Dr. Satyawati Sharma and other faculty members of the centre for their encouragements and valuable suggestions during the course of this work.

I would like to offer special thanks to the former head of the centre, Prof. P. L. Dhar, for his whole-hearted cooperation by making available the necessary experimental and analytical facilities.

I feel very grateful to my institute, IIT Delhi, for enriching my life with wonderful memories and a great learning experience. The financial support provided by the institute is also gratefully acknowledged.

I would also like to thank Gogia Chemical Industries Ltd., Okhla, New Delhi and Mr. Milan Mehta (M/s Minal Exim, Mumbai) for providing reference standards without which the analytical works would not have been possible.

My whole hearted regards and thanks goes to my best friends Dr. Ram Chandra, Dr. Pramod Kumar, Dr. Narendra Kumar Agnihotri, Dr. Ritunesh Kumar, Dr. Madhumita Patel, Dr. Prasanta Kumar Rout, Dr. Lekha Charan Meher and Dr. Parag Bhurchandi for lending me an ear whenever I cried out my heart to them, for all the knowledge sharing and all the fun we have had together and most importantly for keeping my spirit alive and making life a bit colourful during my stay at IIT Delhi.

I am also thankful to Dr. Ashutosh Murkute, Dr. Sophia, Dr. Richa Negi, Kishore Gedam, Lalit Bal, Lalit Joshi, Rama Pradhan, Narendra Sahoo and Rajesh Katiyar for their good company. My special thanks to Mr. Ganesh Prabhu for helping me in analytical works.

I am, indeed, greatly indebted to my family members for their affection, moral support, encouragement and patience they have shown not only during the tenure of this work but also throughout my upbringing to this age. I would also like to thank my in-laws for their patience and motivational support during the last stages of this work.

I express my deepest affection, appreciation and thanks to my wife Rupa for her understanding, patience and sacrifices. Without her constant encouragement and loving support it would have been impossible for me to complete this task.

Last but not the least, I thank all my well wishers who do not figure in this acknowledgement but have helped me in one way or the other during this period.

(Jadeja Girirajsinh Chandrasinh)

ABSTRACT

Of late, there has been a growing shift towards the use of plant-derived molecules (phytoconstituents) in the sectors as diverse as flavourings and fragrances, pharmaceuticals, nutraceuticals, cosmeceuticals, perfumery and even pesticides due to the increased consumer awareness and tighter governmental regulations regarding the health and safety aspects of synthetic chemicals. The most commonly used traditional methods for isolating bioactive phytochemicals from medicinal and aromatic plants; viz. maceration, percolation, steam distillation, hot continuous solvent extraction, decoction, infusion, etc. are time consuming besides using large amounts of toxic organic solvents. Moreover, the continually shrinking list of safe solvents, sky rocketing prices of energy and thermal instability of most of the natural ingredients have given way to some novel extraction technologies such as Supercritical Fluid Extraction (SFE), Pressurized Liquid Extraction (PLE), Ultrasound Assisted Solvent Extraction (UAE) and Microwave Assisted Solvent Extraction (MAE). The need of the hour is, therefore, to design, develop and commercialize 'green' extraction technologies that use no petroleum based solvents at all or comparatively less solvent and that may satisfy all the current as well as foreseeable future regulations concerning safety, health and environment. In accordance, the theme of the present research work was to develop green extraction method to recover high value bioactive components from renewable raw materials.

The present research work was carried out with a focus on developing a green extraction method utilizing pressurized solvents for recovering important phytochemicals from Vanilla (*Vanilla planifolia* Andrews) cured beans and Neem

(*Azadirachta indica* A. Juss) seed kernels. Both the selected plants are source of high value chemical compounds. Vanilla is known for its flavour compounds that are finding increasing use in ice-creams, food and beverages and perfumery whereas Neem is being exploited for extracting limonoid compounds that are widely used in biopesticidal formulations. The operating conditions (temperature, pressure, solvent composition, flow rate, particle size and extraction time) for the optimal yield of the desired phytochemicals from both the plants have been established. Sub- and supercritical CO_2 extraction was also employed for the recovery of natural vanilla flavour. In addition, the extraction efficiency of the proposed method was also compared with their conventional counterparts. Mass transfer studies involving mathematical modeling of the experimental data were performed to provide useful information for process scale-up and yield predictions in the commercial scale facilities. Lastly, an attempt was also made to carry out preliminary economic analysis of a small scale industrial unit based on the proposed method.

The Pressurized Liquid Extraction (PLE) of both the plants revealed that pressure had an almost insignificant effect on the extraction efficiency. However, an enhanced vanilla oleoresin yield with an improved flavour profile was obtained with an increase in temperature. 40% aqueous ethanol was the best solvent for maximizing vanilla oleoresin yield but it was 70% aqueous ethanol which resulted in oleoresin of best flavour profile with maximum vanillin content at a temperature of 90 °C. Apart from vanillin, the other major flavour compounds extracted were p-Hydroxybenzaldehyde, vanillic acid and vanillyl alcohol. In case of PLE of defatted neem kernels, a temperature of 50 °C and a pressure of 50 bar resulted in maximum azadirachtin recovery. The best particle size was found to be 60 mesh. Higher temperatures caused thermal degradation of azadirachtin. Mathematical analysis of the experimental data showed that spherical model (in case of PLE of neem kernels) and flake model (in case of vanilla beans) can be applied to predict the time required to obtain a given extraction yield and thereby rendering useful process scale-up process. The economic appraisal revealed that a small-scale processing unit based on the developed PLE method is commercially viable for both the selected plants. The return on investment and pay-back period for vanilla extraction unit (360 kg/day capacity) were found to be ~20% and ~3 years, respectively. For the neem extraction unit (2 MT/day capacity), these were ~25% and ~2.5 years, respectively. Establishment of such units in the regions with easy and sustained availability of these raw materials will help in their value addition and thereby giving boost to the rural economy by providing better returns to the growers and by additional employment generation.

As compared to the conventional methods, PLE was found to be superior resulting in higher yields of desired phytochemicals in shorter duration (1 h) with a 1.5 to 5 fold reduction in solvent consumption. This, in turn, would pave the way for green alternative method for recovering bio-based products from a gamut of renewable raw materials by reducing environmental impact of bulk organic solvents utilization in traditional extractions.

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