An improved method for the recovery of essential oil from Celery seeds

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Received 25 February 2002; revised received 3 January 2003; accepted 7 April 2003

**Celery**, *Apium graveolens* L. N.O. Umbelliferae is mostly cultivated in Punjab, Haryana, and U.P. for its seeds. The seeds are used as spice to flavour soups, salad, tomato juice, sauces and for extracting essential oil or oleoresins. Of late, they have attained ever increasing popularity among food products. Indian celery is considered to be best in the world and that is why it is in great demand in European countries. In 1999-2000, 4038 MT of seeds worth Rs 9.5 crores and its oleoresin 146.5 MT worth Rs. 4.5 crores were exported. These are prepared by extracting the celery seeds with volatile solvents like alcohol and driving off the alcohol in vacuo. On steam distillation it yields pale yellow essential oil with characteristic rich long lasting spicy odour, the principal components of which are d-limonene and salinene. The oil is used for flavouring food products, liquors and pharmaceutical preparations. Celery is recommended for rheumatic arthritis, antispasmodic, aphrodisiac, nerve-stimulant and hypertension. It induces menstruation in non-pregnant women. It is better known as a medicinal herb than a garden vegetable. The major phthalides being 3-butyl phthalide, 3-butyl tetrahydrophthalide or sedanolide. Others being 3-butyl-1,4,5-dihydrophthalide or sedananolide. These form high boiling constituents of the oil besides the low boiling monoterpenic hydrocarbons e.g., a-pinene, a-terpinene, y-terpinene, b-pinene, limonene etc.

The methods in vogue for recovery of essential oil from celery seeds is by hydrodistillation or steam distillation. The steam distillation is avoided as it leads to channeling inside the fine mass which results in longer time or low recovery. Mostly hydrodistillation is followed and is adopted here. Fischer¹ found that the yield of oil ranged from 1.3-2.5%, the average being 1.78%. Guenther² described isolation of essential oil of celery seeds by steam distillation. The oil yield varies from 1.9-2.4% in 10-12 h. A recent work carried out by Chowdhry & Gupta³ using hydrodistillation gave an oil yield of 2.2% only with phthalides content 10.5% in 10 h. Hydrodistillation was adopted in the present work, wherein a yield of 2.44-2.68% was achieved in 4-4.5 h and the phthalides content in the oil improved to 23 ± 2% compared to 10-12% in 10-12 h reported above. The novelty of the process lies in the true/scientific utilization of the principle of steam or hydrodistillation. In steam or hydrodistillation, the vapour comprises of steam and the volatile component of oil in the ratio given by the relation.

\[ \frac{m_A}{m_B} = \frac{P_A}{P_B} \times \frac{M_A}{M_B} \]

where \( m_A \) and \( m_B \) are the masses of the oil component \( A \) and steam \( B \) respectively, \( M_A \) and \( M_B \) are the molecular weights of the oil component \( A \) and steam \( B \) and \( P_A \) and \( P_B \) are the vapour pressure of oil component \( A \) and steam \( B \) at the boiling point of the mixture. By applying Phase rule, the system has only one degree of freedom, so that either temperature or the pressure may be fixed. In steam or hydrodistillation the temperature gets fixed, whereas the water and the oil component each exert a partial pressure equal to their respective vapour
pressure at the boiling point of the mixture. However
this relation will be only approximately true if the
material is slightly miscible with water. At constant
temperature the above relation can also be put as under:

\[ \frac{m_A}{m_B} = \frac{M_A}{M_B} \times \frac{P_{\text{Al}}}{P_{\text{A}}} \]

or

\[ \frac{m_B}{m_A} = \frac{M_B}{M_A} \times \frac{P_{\text{Al}}}{P_{\text{A}}} \]

where \( P \) is the total pressure of the system.

This shows that mass ratio of water to oil component in the vapour will increase with increase
in molecular weight of oil component or decrease in vapour pressure or volatility of oil component. This
principle has been made use of, in the isolation of phthalides from celery seeds. Thus by increasing the
mass ratio of steam to celery seeds or in other words to celery seed oil, matching with partial pressure of
phthalides, it has been possible to get maximum oil/phthalides in minimum period of time. The
distillation was stopped where cost of steam versus oil yield made the process uneconomical. The novelty
lies in visualizing, working out and utilizing the principle involved in the recovery of high boiling
components present in the oils of natural products such as celery seeds, \( Cymbopogon flexuosus \) grass,
etc., from which the oil is recovered by steam/hydrodistillation.

Today tons of celery seed is being steam/hydrodistilled in India and abroad getting low
yield with low content of high boilers like phthalides, which are the main valuable components and the time
taken for distillation is around 3 times the desired time. This idea will enlighten the distillers to make
use of the principle in all cases, thereby improving economics wherever possible. Without this lot of
valuable oil is not being recovered and lost.

**Experimental Procedure**

**Plant material**

Celery seeds were procured from Ms. Suran Singh
Ram Singh, Maqboolpura, Amritsar, Punjab (India).

**Isolation of essential oil**

Celery seeds were crushed to half the seed size in
household grinder and hydrodistilled with a known quantity of water, 3 to 5 times the weight of the seeds,
using (i) clevenger (ii) straight distillation. Time of
distillation was varied from 3 to 5 h, at the boiling
point of water. The experiments were conducted
with/without the addition of NaCl and repeated under
microwave irradiation. The residue left was dried and
used for extracting fat and as boiler fuel. The oil
obtained was analysed by GLC and its phthalides
identified by GC/MS.

**Results and Discussion**

The work was initially carried out in
clevenger/straight distillation using thermal heating.
The volume of the clevenger triangle as well as the
circulation rate \( \text{mL/h} \) was found to affect the oil yield as observed in Table 1.

In case of clevenger, the circulation rate was determined by observing the time of filling triangle in
seconds. It was found that in case of clevenger with small triangle, volume and higher circulation rate, the
oil obtained was less which was due to low retention
time of distillate which did not give enough time for
the separation of oil. This rate was optimised and
found to be double (1.9 times) as per Sl. No. 4
Table 1, which gave a maximum yield of 2.2% oil
(\( \text{v/w} \)), containing 22.9% phthalides. To overcome the
difficulty of retention time, experiments were
repeated using straight distillation and passing the aqueous distillate through \( n \)-hexane before recycling.
This improved the yield to 2.66% with phthalide
content of 25% (Sl. No. 6, Table 1).

Under similar circulation rate, addition of salt
decreased the oil yield even with the same time of
distillation (Sl. No. 5, Table 1), which showed
negative effect of salt, probably due to rise in boiling
point and polymerization. The experiments were
repeated under microwave irradiation using similar
parameters (Table 1).

It may be observed that the yield of the oil is comparatively less by 2% under conditions shown in
Table 1 at Sl. No. 7 as compared to experimental
conditions at Sl. No. 4, Table 1, though the circulation
rate was on the higher side. This was further
confirmed from experiment at Sl. No. 10 in Table 1,
where salt was used. The yield of the oil was 2.5%
compared to 2.66% from experiment Sl. No. 6
Table 1, under parallel conditions. The reason for low
oil yield under microwave irradiation conditions was high h.p. of the solvent.

Studies were scaled up on the pilot plant scale
(Table 2) using stainless steel distillation still (steam
jacketed) of 50 litres capacity. It was observed that
the oil yield under optimum conditions was 2.44% in 4 h and 2.68% in 4.30 h. Thus, it may be concluded that distillation rate per unit weight of the seeds is important and should be kept around two for maximum yield and optimum distillation time 4-4.30 h. Thus, it may be further concluded that the phthalides content increases with the circulation rate which is necessary for bringing out the high boiling distillates.

Acknowledgements
The authors are thankful to Director, RRL Jammu for his keen interest in this project.

References
1. Fischer T, Bull Nail Farm Comm, 13 (1945) 6.