Sulphur terpenoid concentration by counter-current fractionation: A feasibility study

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ABSTRACT

Buchu (*Agathosma crenulata*) is a species of a shrub that is indigenous to the southern and western parts of the Western Cape Province of South Africa. It is valued for its characteristic fruity-minty-herbal smell, as well as its proven therapeutic properties. The essential oil (EO) obtained from its leaves is highly valued. It is made up mainly of monoterpene hydrocarbon compounds (MT) and their oxygenated derivatives (OT), and, in much smaller quantities, some especially highly valued sulphur derivatives of monoterpene hydrocarbons (ST), which find use in the flavor and fragrance industries. Separation of the high value compounds from the terpenes and their derivatives is essential, as monoterpenes are prone to oxidation upon exposure to air. Pulegone, an oxygenated monoterpenoid present in high concentration in Buchu essential oil, is a hepatotoxin, and has an especially overpowering malodorous earthy-minty smell, and is thus undesirable in the product.

Information on the separation of ST from OT and MT using any method of separation could not be found in the literature. Vacuum distillation and solvent extraction are some of the traditional methods of fraction of EO components. Thermal methods, however, result in the decomposition of the often thermally labile high value compounds, while organic solvents tend to persist in the product, and thus adversely affect the quality of the product.

This work investigates the feasibility of separation of ST from MT and OT using supercritical CO_2 (sCO_2). Phase behaviour data for the binary systems of the main components of the EO could not be found in the literature, and were therefore measured. The data were fitted to the RK-Aspen equation of state for conditions between 308 K and 358 K, and 0.011 and 0.65 solute mass fractions. The calculated distribution coefficients and separation factors for a simulated feed indicated a high selectivity for the separation ST/MT, and modest selectivity for the MT/MT separation. The separation ST/OT was predicted to be more difficult. The data also provided the range of pressure and temperature conditions for a successful separation.

Using the phase equilibria study results, a process model was developed using the commercial process simulator Aspen Plus®, and validated using pilot plant data. Various scenarios of fractionation in a counter-current column were investigated. The results indicated that while deterpenation (MT/OT) could be achieved in a single stage separation, the OT/ST could not be achieved satisfactorily. However, a two stage separation could successful separate MT/OT/ST, on condition that the first stage of separation was optimized for the MT/OT separation. It is thus possible to separate ST from MT and OT using sCO_2.