Supercritical Carbon Dioxide Extraction of Andrographolide from Andrographis paniculata: Effect of the Solvent Flow Rate, Pressure, and Temperature*

A.C. Kumoro^a and Masitah Hasan^{b,**}

^a Department of Chemical Engineering, Faculty of Engineering, Diponegoro University, SH Road, Tembalang Semarang 50139, Indonesia

Abstract Andrographis paniculata Nees has been extensively used for traditional medicine and help against fever, dysentery, diarrhoea, inflammation, and sore throat. In this study, andrographolide, the main component of this plant was extracted from the leaves of A. paniculata using supercritical carbon dioxide. The operating pressures were varied from 7.50 to 20MPa, the temperatures were varied from 30°C to 60°C, and the flow rates were varied from 0.5 to 4ml·min^{-1} . The best extraction condition occurred at 10MPa, 40° C, and a flow rate of 2ml·min^{-1} for a 3g sample of A. paniculata ground-dried leaves. The measured extraction rate was found to be about 0.0174g of andrographolide per gram of andrographolide present in the leaves per hour of operation. The future studies must focus on the interaction between the various operating parameters such as temperature, pressure, and flow rate of supercritical carbon dioxide.

Keywords supercritical carbon dioxide, extraction, andrographolide, *Andrographis paniculata*

1 INTRODUCTION

Andrographis paniculata Nees, locally known as Hempedu Bumi and commonly called the King of Bitter grows widely in the tropical area of South East Asia, India, and China with a plant height of 30—70cm. In these regions, this plant has been extensively used for traditional medicine and help against fever, dysentery, diarrhoea, inflammation, and sore throat[1]. Furthermore, it is a promising new way for the treatment of several diseases, including HIV, AIDS, and numerous symptoms associated with immune disorders[2].

The three main diterpenoid lactones identified in A. paniculata leaves were andrographolide, neo-andrographolide, and deoxyandrographolide [3,4]. Andrographolide, which is grouped as an unsaturated trihydroxy lactone has the molecular formula of $C_{20}H_{30}O_5$. The molecular structure of andrographolide is shown in Fig.1. Andrographolide, the main component in the leaves of A. paniculata can be easily dissolved in methanol, ethanol, pyridine, acetic acid, and acetone, but slightly dissolved in ether and water. Its physical properties are: melting point at $228-230^{\circ}C$, and the ultraviolet spectrum in ethanol λ_{max} as 223nm[4].

Figure 1 Molecular structure of andrographolide

Conventional production methods such as solvent extraction and Soxhlet, although effective for extraction, can lead to degradation of heat sensitive compounds as well as leave traces of toxic solvents in the solute. This is a concern for food and medicinal extracts[5]. Supercritical fluid extraction may be a viable alternative to solvent extraction methods. The phenomenon of supercritical fluid extraction was recorded over 100 years ago but has been slow in finding commercial applications, partly because of the sophisticated and expensive high pressure equipment and technology required [6].

Carbon dioxide is generally the most desirable solvent for supercritical fluid extraction (SFE). Its low critical temperature (31.26°C) and pressure (7.38MPa) make it effective for the extraction of heat sensitive (thermally labile) compounds. In addition, it is an inert, non-flammable, non-explosive, inexpensive, odourless, colorless clean solvent that results in no solvent residue in the product; it is also non-toxic and is generally accepted as a harmless ingredient in pharmaceuticals and food[5]. It also has low surface tension and viscosity and high diffusivity, which make it attractive as a supercritical solvent[7]. The diffusivity of supercritical carbon dioxide is one to two orders of magnitude higher than for other fluids, which permits rapid mass transfer, resulting in a larger extraction rate than that obtained by conventional liquid extraction[5]. Although several studies about supercritical fluid extraction have been carried out in the last few decades, only one article reported about the supercritical extraction of andrographolide from A. paniculata leaves. Ge et al.[8] reported the use of supercritical carbon dioxide as well as carbon dioxide and ethanol mixture to extract andrographolide in an own built extractor

^b Department of Chemical Engineering, Faculty of Engineering, University of Malaya, Kuala Lumpur 50603, Malaysia

Received 2006-09-29, accepted 2007-08-29.

^{*} Supported by the Intensification of Research in Priority Areas Project (IRPA), Ministry of Science, Technology and Innovation, Malaysia (No.09-02-03-0101-EA0001).

^{**} To whom correspondence should be addressed. E-mail: masitahhasan@um.edu.my