## Asymptotic solution for the model of supercritical fluid extraction from polydisperse ensemble of ground particles with high oil content

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The process of supercritical fluid extraction (SFE) employs fluids to extract vegetable oils from plant material. The extraction from ground seeds with high oil content is studied. A representative particle ensemble is demonstrated in Figure 1c. It is placed in the cylindrical column of the extractor before the experiment to form a porous packed bed as shown in Figure 1a. The solvent is pumped through the packed bed at a fixed rate. It penetrates the raw material and dissolves the oil which diffuses to the particle surface. Then, the filtrating solvent transports the solute to the extractor column outlet cross-section.

Established modelling approach suggests the use of a monodisperse spherical approximation of the particle ensemble with the characteristic particle radius  $a_0 \sim 500 \,\mu$ m. However, recent experimental observations revealed that the size distribution is essentially bimodal: two fractions, or modes, has to be distinguished. While the first modal value,  $a_0$ , corresponds to the regular particles, the second mode describes the so-called "dust" particles with the characteristic radius  $a_1 \sim 50 \,\mu$ m, as seen in Figure 1c. The dust particles are always present in the packed bed of ground oil seeds.

The SFE process is considered as a heterogeneous surface reaction, and the density of the particle size volume distribution, f(a) = dF/da, is one of the major parameters. The sensitivity of the Overall Extraction Curve (OEC), Y(t) (where t is time), to f is theoretically analysed, and the minimum set of parameters that is sufficient to characterize the polydispersity of packed bed is suggested. First, we extend the differential mass balance equations of the SFE to take into account the polydisperse nature of the ground particles. Then, we derive the closed form expression for Y(t) that is treated asymptotically. The density function is presented as a combination,  $f(a) = \alpha f_1(a) + (1-\alpha)f_2(a)$ , of two density functions, where  $\alpha$  is the volume fraction of the dust particles,  $f_1(a)$  is the density of their size distribution, and  $f_2(a)$  is the density of distribution function corresponding to the regular particles. The value of  $f_2$  argument scales as  $a_0$  while  $a_1 << a_0$  is the scale for the  $f_1$  argument.  $1/a_0$  is the small parameter of the asymptotic expansion. The accuracy of the approximate solution is demonstrated in Figure 1d.



**Figure 1. (A)** A porous packed bed of ground spherical particles in the cylindrical column of height *L*. The direction of the solvent flow is along the *z*-axis. **(B)** Schematics of mass transfer in a single spherical particle of radius *a* within the framework of the shrinking core approach. Two sub-zones are distinguished: (1) internal oil-containing core of radius *R*, (2) external transport zone where the oil is depleted. Wavy arrows – diffusion paths of dissolved solute against the concentration gradient. The core shrinks with time *t* as shown by dashed circles and the arrow. **(C)** Bimodal nature of ground particles size distribution. Scale: 800  $\mu$ m. **(D)** A set of normalized OECs *Y* vs. dimensionless time  $\tau$  at different volume fractions  $\alpha$  of dust particles. Solid curves – exact solution of the problem, dashed curves – asymptotic expansion: black – two terms, red – improved expansion.  $\tau$  – dimensionless time.

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