Simulation of Supercritical Fluid Extraction Process Using COMSOL Multiphysics

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Abstract: The present work deals with the simulation of mathematical model for supercritical extraction process of Sage leaves . Reverchon,1996 extracted sage oil using supercritical extraction method from sage leaves at 9 MPa and 50 ℃. Different hypotheses were tested on vegetable matter geometry, and their incidence on the model performance was evaluated. The model was also developed by Reverchon, 1996. Four mean size of sage leaves ranging from 0.25 to 3.10 mm were taken for extraction with other experimental conditions and process parameters. Experimental results were fitted in the model and the results obtained were in a good agreement. In present work, same mathematical model is solved using COMSOL multiphysics and results are compared with the results given in literature to find the utilisation of COMSOL multiphysics.

Keywords: Supercritical fluid extraction, Sage leaves, Equation based modeling

1. Introduction

Supercritical extraction process is promising and benign alternative to extract high value added products for the food, cosmetics, and pharmaceutical industries. The fluid above this critical temperature and pressure is called a supercritical fluid. Supercritical fluid extraction is a technique that exploits the solvent power of supercritical fluids at temperatures and pressures near the critical point. With supercritical fluid extraction (SFE) higher yields and better quality products can be achieved. Carbon di-oxide at its supercritical conditions (SC-CO_2), is the most desirable solvent for the extraction of natural products as it is non-toxic, inexpensive, non-flammable, and non-polluting. SC-CO_2 is used in food applications as a solvent for the extraction of non-polar solutes. Supercritical extraction method is used for the extraction of several natural products such as: sunflower seed, watermelon seed, black pepper seed, rosemary flower, ginger root, turmeric root etc.

2. Literature Review

Several mathematical models on supercritical fluid extraction process have been proposed by different authors as given in literature. Reverchon, 1996 used sage leaves for the extraction of sage oil using supercritical extraction method at 9 MPa and 50 ℃. The mean particle size was evaluated by mechanical sieving. Batches of 3.10, 0.75, 0.50 and 0.25 mm mean particle sizes were used for the extraction. A mathematical model based on differential mass balances performed along the extraction bed was also developed by Reverchon, 1996 and used to fit experimental data of sage leaves. Other experimental conditions and process parameters used for the extraction are shown in table 1.

Table-1 Experimental conditions and process parameters

Parameter	Value
Flow rate (Q)	8.83 g/min
Porosity (ε)	0.4
Superficial viscosity	$0.455*10^{-3}$ m/s
Fluid phase coefficient (k_f)	$1.91*10^{-5}$ m/s
Amount of seed (W)	0.160 kg
Volumetric partition coefficient (k_p)	0.2
Diffusivity (D_i)	$6.0*10^{-13}$ m^2/s

3. Model development

Proposed model was based on differential mass balances performed along the extraction bed (fig. 1) while neglecting external mass transfer coefficient and axial dispersion in addition to the constant solvent density and flow rate along the bed (Reverchon,1996). The mass balances over an element of the extractor of height *dh* can be written as:

$$uV\frac{\partial c}{\partial h} + \varepsilon V\frac{\partial c}{\partial t} + A_p K(q - q^*) = 0 \qquad \text{(i)}$$

$$(1-\varepsilon)V\frac{\partial q}{\partial t} = -A_p K(q - q^*) \qquad \text{(ii)}$$

With initial and boundary condition as:

$$c = 0, \quad q = q_0 \quad \text{at } t = 0$$

(iii)

$$c(0, t) = 0 \quad \text{at} \quad h = 0$$

A linear relationship was used due to lack of experimental data:

$$c = k_p \cdot q^*$$

(iv)

In equation (ii), $\dfrac{A_p K}{(1-\varepsilon)V}$ depends on the geometry of particles and dimensionally equal to the 1/s, thus::

$$t_i = \frac{A_p K}{(1-\varepsilon)V} = \mu \frac{l^2}{D_i}$$

(v)

For spherical particle μ is equal to 3/5 and l is equal to V_p/A_p (particle volume/particle surface).

Super critical fluid and
extracted solute

Z=h

$C_{z+\Delta z}$

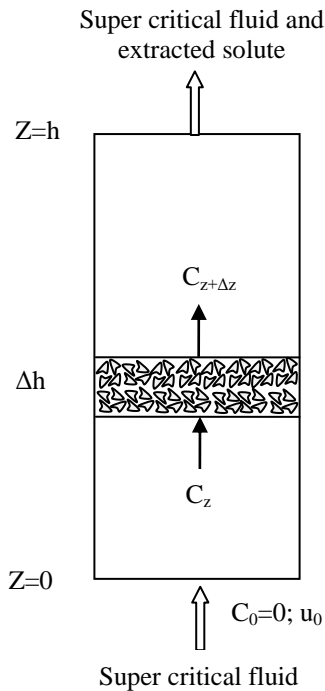Δh

C_z

Z=0

$C_0 = 0; u_0$

Super critical fluid

Figure 1: Schematic representation of extraction bed

Authors solved the model by rewriting mass balance equations as a set of 2n equations as given in eq. (vi) and (vii) and solved numerically using fourth order Rungee - Kutta method.

$$\frac{W}{\rho}(c_n - c_{n-1}) + \varepsilon \frac{V}{n} \frac{dc_n}{dt} + (1-\varepsilon)\frac{V}{n}\frac{dq_n}{dt} = 0$$

(vi)

$$\frac{dq_n}{dt} = -\frac{1}{t_i}(q_n - q_n^*)$$

(vii)

4. Model solved by using COMSOL multiphysics

The model proposed by Reverchon, 1996, as explained above, is solved using COMSOL multiphysics 5.0 software in the present work. Equation based modeling approach is used to solve the model equations instead of any built-in physics interface like Chemical Spices Transport Physics Interface. It is recommended to use built-in physics interfaces to enable ready-made post-processing variables and other tools for faster model setup with much lower risk of human error. To solve the model equations (i) and (ii), two interfaces were taken under Mathematics branch: (i) PDE interfaces and (2) ODE and DAE interfaces. For equation (i), **The Coefficient form PDE interface** is taken because it contains two independent variable 't 'and 'h' and **Domain ODEs and DAEs** is used for equation (ii) because of presence single independent variable 't'. For the solution, 1D geometry is taken to simplify the problem. Time dependent study option is taken after choosing two different interfaces under Mathematics branch.

5. Use of COMSOL multiphysics

Model equations are solved considering 1D geometry under Model wizard window on COMSOL desktop. Under Select Physics option, Mathematics interface group is selected and then Coefficient form PDE under PDE subgroup and global ODEs and DAEs interface under ODE and DAE subgroup are added. After adding physics, Time dependent study was carried out as both models equations are differentiated with respect to time 't'. While entering Interval values (Left endpoint and Right endpoint) under Geometry option in Model builder window and a line is formed as 1D geometry as shown in Fig. A-1 and A-2. Some parameters used in the process are constant and their values are added in parameters section in Model builder window as given in Fig. A-3. Two dependent variable (solvent phase oil concentration 'c' and solid phase oil concentration 'q') and two independent variables (time 't' and height of bed 'h') are considered for solving the equations. Other than these variables, a variable is added by right clicking the Definitions option under Component 1 section in Model builder window and

can be seen in Fig. A-4. Further, Equation (1) is compared with the default equation of Coefficient form PDE and values of coefficients are added in the Coefficient form PDE 1 window as shown in Fig. A-5. Initial value is added with help of pointwise constraint setting because as given in eq. (iii), value of dependent variable 'c' is zero at h=0 and can be noticed through Fig. A-6 and A-7. For applying pointwise constraint setting Left endpoint is considered as h=0 (after which extraction starts) and Right endpoint is considered as Z= h, height of column (extraction ends) as given in figure 1. Similarly eq. (ii) is compared with the default equation given in Distributed ODE 1 interface and value of coefficients are added as shown in Fig. A-8. Initial conditions are added for this equation as given in eq. (iii) in Initial values section under Domain ODEs and DAEs interface and can be seen by Fig. A-9. This study is time dependent study so a range for time value should also be given for the solution of equations and is shown in Fig. A-10. Under study setting, time unit, time range and tolerance values are added and then model is computed to get the results as shown in Fig. A-11.

6. Results and Discussions

Results by the computation of the model are compared with the results given in the literature. Results are computed considering four different particle size and plotted between extraction yield (amount of oil extracted*100/amount of oil available in seed) and extraction time. As given in literature, Extraction yield values for 0.25 and 0.5 mm particle size are 100% as can be seen from Fig. 2 and shows that almost total amount of oil available are extracted by the extraction process. A lesser extraction yield value is observed for 0.75 and 3.1 mm particle, which was 90% and 20%, respectively. Comparison of results from literature (Reverchon,1996) and COMSOL multiphysics are given in figure (2) and figure (3), respectively. While comparing with the results from COMSOL multiphysics, extraction yield results for 0.25, 0.5 and 0.75 mm particle size are same as found in literature. However, for the particle size 3.1, extraction yield observed is lesser than that of literature. The reason for the less extraction yield could be that model is not supporting the extraction of large particle size solute. Computation time taken by the COMSOL multiphysics is 12 s which is far lesser than other software packages and larger memory usage also requires for Runge- kutta method which is used in literature to solve the model equations.
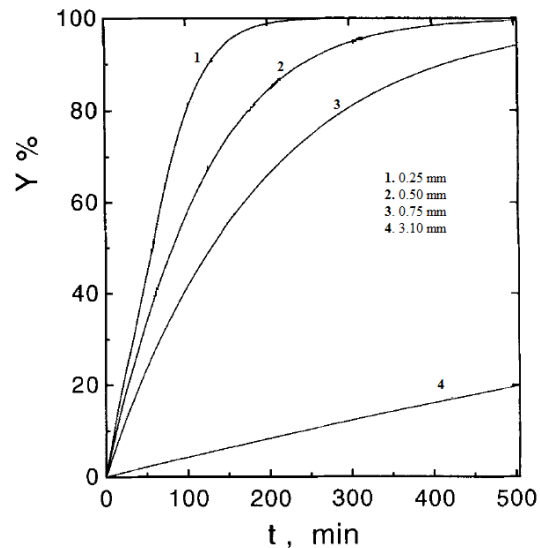

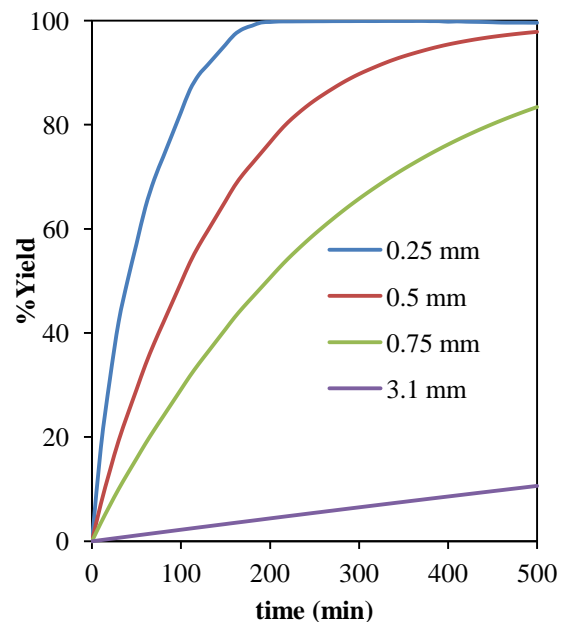Figure:2 Results given by Reverchon,1996

Figure:3 Results from COMSOL multiphysics 5.0

7. References

1. E. Reverchon, Mathematical Modeling of Supercritical Extraction of Sage Oil, *Bioengineering, Food, and Natural Products*, **Vol. 42**, No. 6 (1996).

2. Eduardo L.G. Oliveira et al., Review of kinetic models for supercritical fluid extraction, *chemical engineering research and design*, **89**,1104-1117 (2011).

3. Mamata Mukhopadhyay, Extraction and processing with supercritical fluids, *J Chem Technol Biotechnol*, **84**, 6-12 (2008).
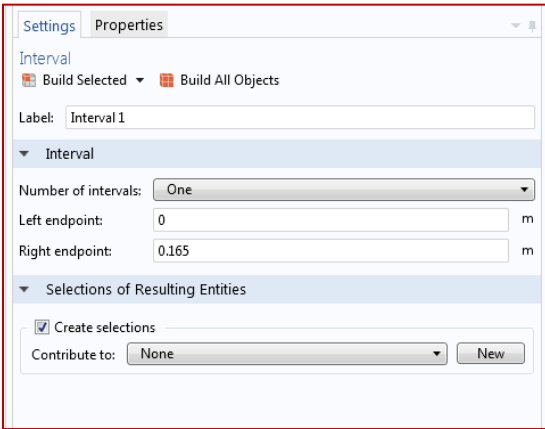
10. Appendix

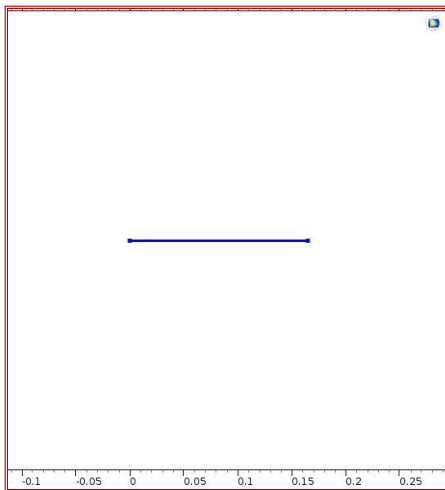Figure A-1 Geometry setting window for the model.
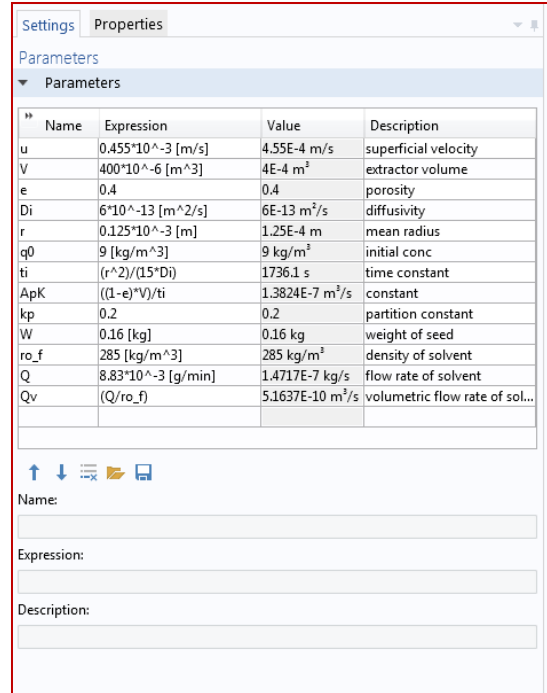

Figure A-2 1D geometry window for the model.
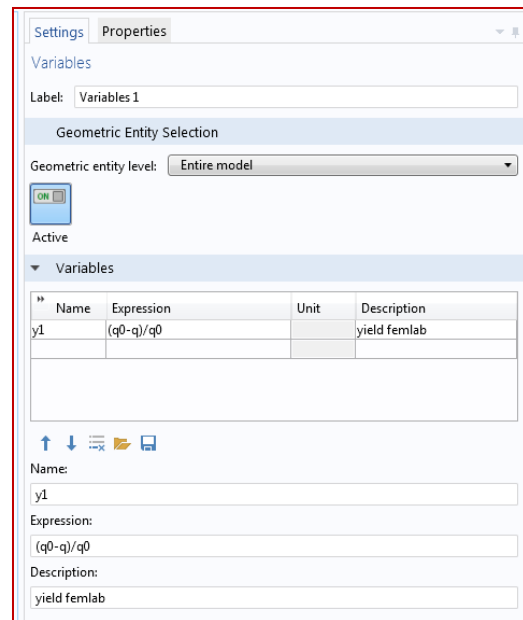

Figure A-3 Parameters setting window of the model.

Figure A-4 Variable setting window of the model.
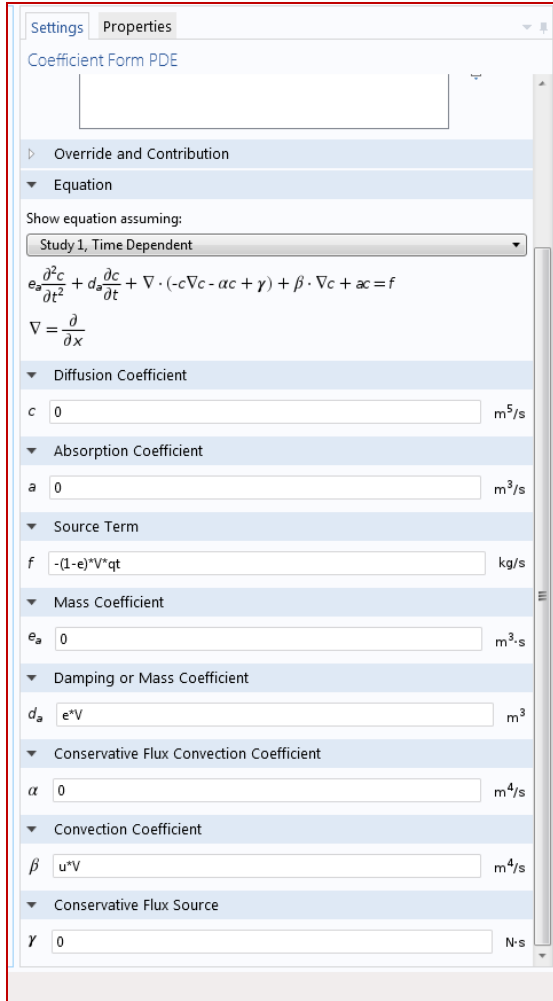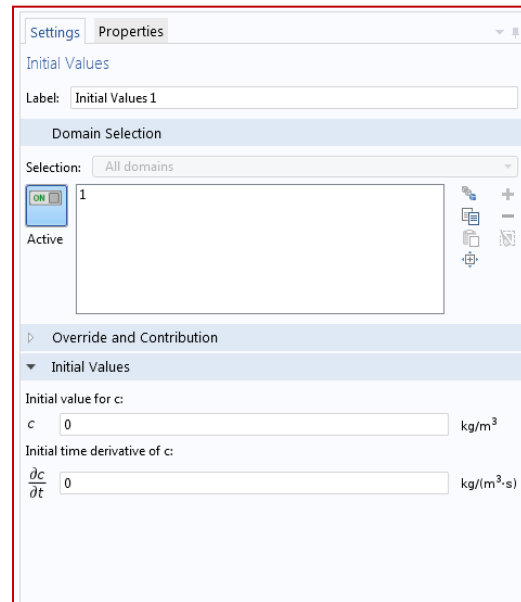
Figure A-5 (Coefficient Form PDE)

Settings | Properties

Coefficient Form PDE

▷ Override and Contribution

▼ Equation

Show equation assuming:

Study 1, Time Dependent

$$e_a \frac{\partial^2 c}{\partial t^2} + d_a \frac{\partial c}{\partial t} + \nabla \cdot (-c\nabla c - \alpha c + \gamma) + \beta \cdot \nabla c + ac = f$$

$$\nabla = \frac{\partial}{\partial x}$$

▼ Diffusion Coefficient

c 0 m^5/s

▼ Absorption Coefficient

a 0 m^3/s

▼ Source Term

f -(1-e)*V*qt   kg/s

▼ Mass Coefficient

e_a 0 $m^3 \cdot s$

▼ Damping or Mass Coefficient

d_a e*V m^3

▼ Conservative Flux Convection Coefficient

α 0 m^4/s

▼ Convection Coefficient

β u*V m^4/s

▼ Conservative Flux Source

γ 0 $N \cdot s$

Figure A-5 Coefficient form PDE setting window for equation (i).

Figure A-6 (Pointwise Constraint)

Pointwise Constraint

Label: Pointwise Constraint 2

Boundary Selection

Selection: Manual

Active   1

▷ Override and Contribution

▼ Pointwise Constraint

Apply reaction terms on:

All physics (symmetric)

Constraint expression:

c

Constraint method:

Elemental

▷ Discretization

Figure A-6 Constraint setting window for the boundary condition (ii).

Figure A-7 (Initial Values)

Settings | Properties

Initial Values

Label: Initial Values 1

Domain Selection

Selection: All domains

Active   1

▷ Override and Contribution

▼ Initial Values

Initial value for c:

c 0 kg/m^3

Initial time derivative of c:

$\frac{\partial c}{\partial t}$ 0 $kg/(m^3 \cdot s)$

Figure A-7 Initial values setting window for Coefficient form PDE.

Figure A-8 (Distributed ODE)

Settings | Properties

Distributed ODE

Label: Distributed ODE 1

Domain Selection

Selection: All domains

Active   1

▷ Override and Contribution

▼ Equation

Show equation assuming:

Study 1, Time Dependent

$$e_a \frac{\partial^2 q}{\partial t^2} + d_a \frac{\partial q}{\partial t} = f$$

▼ Source Term

f -ApK*(q - c/kp)   kg/s

▼ Damping or Mass Coefficient

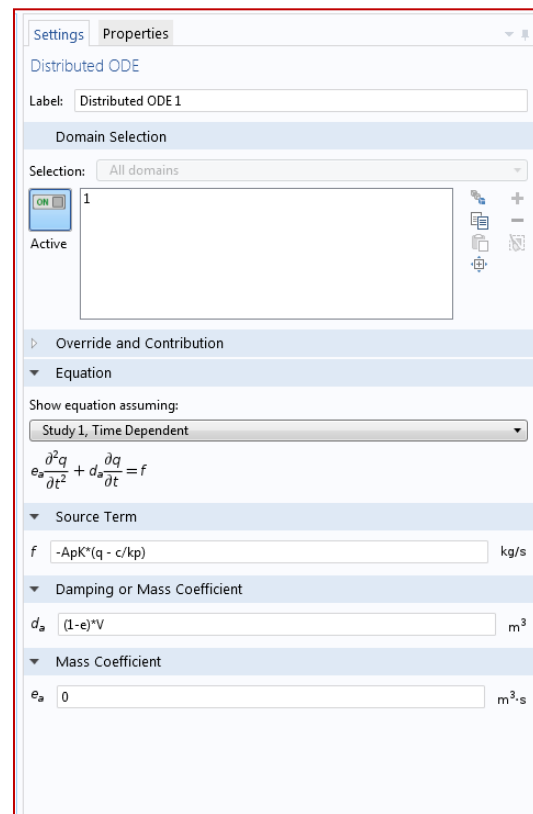d_a (1-e)*V m^3

▼ Mass Coefficient

e_a 0 $m^3 \cdot s$

Figure A-8 Distributed ODE 1 setting window for equation (ii).
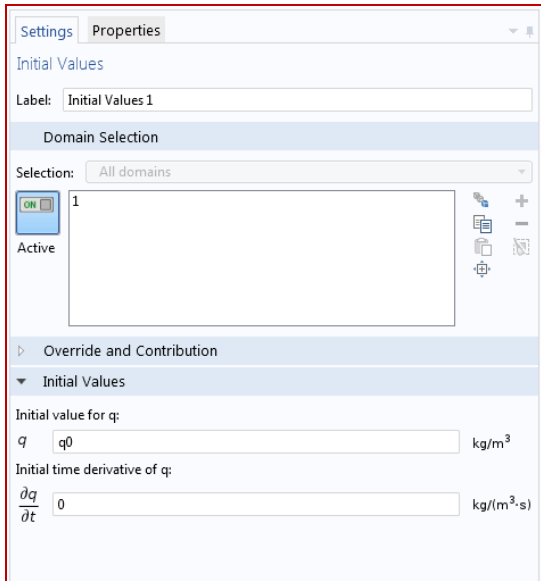
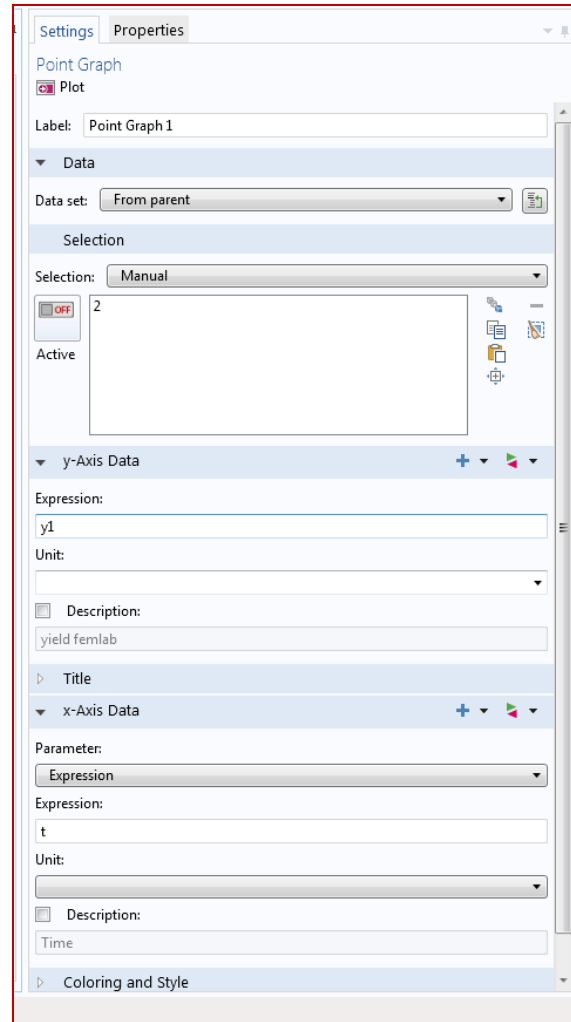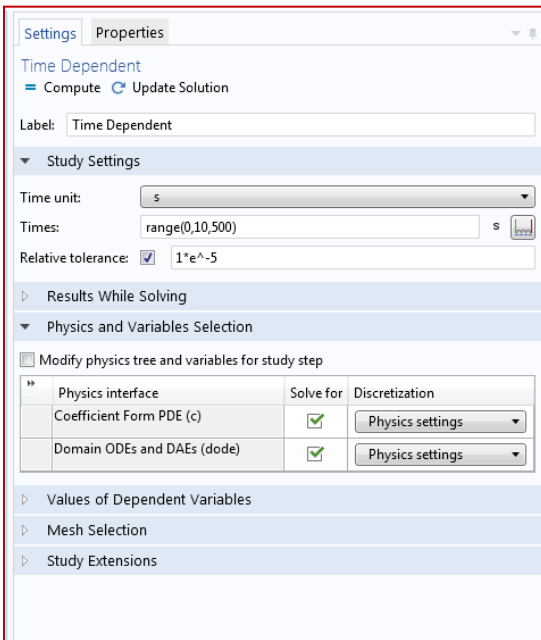Figure A-9 Initial values setting window for Distributed ODE 1.

Figure A-10 Time dependent Study setting for the model.

Figure A-11 Results setting window of the model.