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SIMULATION OF FREE ENERGY OF MIXING FOR A POLYMER SOLUTION USING A SPREADSHEET FOR LEARNING ACTIVITIES

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Abstract. Teaching physics is sometimes difficult to convey without using props or visualizations. Simulations can help teachers create and demonstrate real conditions in front of the class. In this article, we describe the usage of spreadsheets in simulating Gibbs free energy in mixing polymer solutions. We have created an model for the simulation consisting of a main spreadsheet and several secondary spreadsheets. A spreadsheet was chosen to simulate Gibbs free energy because spreadsheets can perform numerical representations in tables. This simple simulation can be used when discussing the topic of polymer thermodynamics. Teachers can start by deriving mathematical equations and then show simulations to visualize the equations. Another option is for students to be asked to create their simulations after deriving a mathematical equation. Using simulation in learning can make learning more interactive and help students understand material physics subjects more easily. Spreadsheets can be an alternative for teachers when explaining abstract material to students. Furthermore, simulations with spreadsheets can also support physics learning remotely.

Keywords: *Simulation, Spreadsheet, Free energy.*

INTRODUCTION

Teaching physics is sometimes difficult to convey without using props or visualizations. Simulations can help teachers create and demonstrate real conditions in front of the class. Simulation can be expected to be an effective tool for helping students to explore their higher level of learning abilities such as analysis, synthesis, and evaluation (Eso et al., 2018). Physics is one of the hard and boring subjects because the material is abstract and difficult to feel. Several education experts develop learning media in the learning process to help teachers and students. Lots of media developed by educational experts both real and virtual (Perdana et al., 2019). Physics as a collection of concepts that explain natural phenomena, even more abstract, complex and difficult for students to understand, needs to be visualized and demonstrated clearly and interactively to create great motivation in learning physics. (Eso, 2018). It can be said that simulations can help teachers explain abstract physics material so that students can understand it more easily.

One of the most important is knowing the appropriate device to maximize the potential of students (M. Oliveira & Nápoles, 2017). Many media on the web or textbooks are usually presented in such a way that it is difficult for students to modify them easily and are presented in languages such as Matlab which are not available to everyone (Robinson & Jovanoski, 2011). It can be difficult for both teachers and students if the device is not easy to use. Although it can provide many benefits, teachers tend to avoid using instructional media when they are difficult to use (Perdana et al., 2019). Spreadsheets are computational and graphical tools that are very useful for creating simulations and visualizations (E. Pratidhina et al., 2020), can help with electronic assessments (Azevedo & Pedrosa, 2017) and provide easy and simple calculation tools (Sokolova et al., 2018). Spreadsheets can be used to process values and create research reports with graphs and tables (Irvan et al., 2021). Spreadsheets can also be used to support the implementation of distance physics learning (Putranta et al., 2022). Currently, spreadsheets provide tools that allow the development of automated procedures such as obtaining animation effects using very simple programming commands (M. C. Oliveira & Nápoles, 2010). The curve generated by the spreadsheet is smoother, dynamic, and can be modified according to the given value (Putranta & Kuswanto, 2018). The usefulness of a spreadsheet as a teaching/learning tool lies in its simplicity and user-friendliness, built-in graphical capabilities, flexibility in changing model parameters, universal availability, ability to be used as or in combination with a word processor (Raisal et al., 2022). The use of graphs in spreadsheets can make learning interactive (Grigore et al., 2016).

Thermodynamics is often considered a difficult subject where many students never feel fully comfortable due to the many equations that are not understood (Del Mar Olaya et al., 2010). So there is a lot of literature suggesting various solutions for an efficient approach to this physics domain (Grigore et al., 2015). Several studies show that students' understanding of the concepts of entropy and Gibbs free energy at several universities is still weak (Carson & Watson, 2002). The concept of Gibbs free energy is very abstract for students and difficult to define. Students often ignore the entropy change of a reaction, and associate exothermic reactions with spontaneity when determining the Gibbs free energy change (Finkenstaedt-Quinn et al., 2020). In this article, we describe the usage of spreadsheets in simulating Gibbs free energy in mixing polymer solutions. This simulation is expected able to be an alternative solution for teachers when explaining abstract material in physics learning.

METHOD

Gibbs free energy is defined as the amount of energy required for a system at a constant temperature with negligible volume, minus the energy transferred to the surroundings due to heat flux (Wilberforce et al., 2017). Gibbs free energy is the most appropriate criterion to evaluate the spontaneity of a process (Barros et al., 2016). Gibbs energy has been used to analyze the formation of ethanol vapor and calculate the theoretical maximum stress of the proton exchange membrane fuel cell (Lazar et al., 2019). The Gibbs free energy of mixing for a system of binary components is shown in equation 1.

$$\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix} \tag{1}$$

Where ΔG_{mix} is the Gibbs free energy, ΔH_{mix} is the enthalpy of mixing for the polymer solution, T is the temperature, and ΔS_{mix} is the entropy of mixing. The entropy equation of mixing is shown in the equation 2.

$$\Delta S_{mix} = -R\left(n_A \ln X_A + n_B \ln X_B\right) \tag{2}$$

Where n_A and n_B are the number of moles of the two components, X_A and X_B are the mole fractions of the two components in the mixture, and R is the gas constant. In addition, the entropy of the mixture can also be calculated using the equation 3.

$$\Delta S_{mix} = -k_B\left(N_A \ln v_A + N_B \ln v_B\right) \tag{3}$$

Where k_B is the Boltzman constant, N_A is the number of solvent molecules, v_A is the volume fraction of the solvent, N_B is the number of polymer molecules, and v_B is the volume fraction of the polymer.

The enthalpy of mixing for an ordinary polymer-solvent solution is

$$\Delta H_{mix} = -k_B T \chi N_A v_B \tag{4}$$

Where χ is the parameter of the Flory-Huggins interaction. The interaction parameters are an important feature of polymer solution theory. This is an indication of how well the polymer interacts with the solvent.

$$\chi = \chi_u + \frac{\eta p}{k_B T} \tag{5}$$

Where η is a parameter similar to χ_u, and p is pressure. Empirically it is known that χ_u is often a linear function of 1/T:

$$\chi_u = A + \frac{B}{T} \tag{6}$$

In some cases, χ_u is a nonmonotonic function of 1/T. In this case, we can use the equation 7.

$$\chi_u = A + \frac{B}{T} + \frac{C}{T^2} \tag{7}$$

So, the complete equation of χ is:

$$\chi = A + \frac{B}{T} + \frac{C}{T^2} + D\frac{p}{T} \tag{8}$$

Where D = $\frac{\eta}{k_B}$. Parameters A, B, C, and D were obtained from fitting experimental data (Knychała et al., 2017). Although χ values can be calculated experimentally, in some cases, predicting χ values can be very challenging. Accurate predictions of the value of χ can only be obtained from precise liquid-like polymer structures (Zhang et al., 2017).

Equations 3 and 4 are substituted in equation 1 to produce equation 9.

$$\begin{aligned}
\Delta G_{mix} &= k_B T \chi N_A v_B - T\left[-k_B\left(N_A \ln v_A + N_B \ln v_B\right)\right] \\
\Delta G_{mix} &= k_B T \chi N_A v_B + k_B T\left(N_A \ln v_A + N_B \ln v_B\right) \\
\Delta G_{mix} &= k_B T\left(\chi N_A v_B + N_A \ln v_A + N_B \ln v_B\right) \qquad (9)
\end{aligned}$$

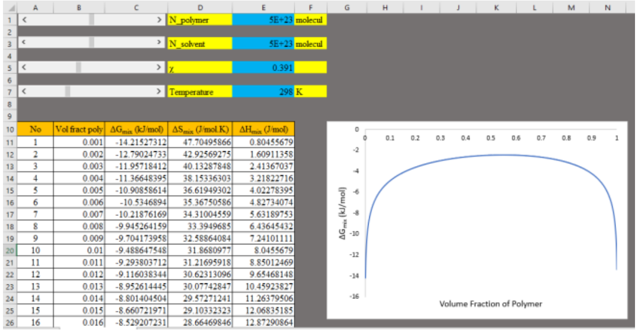
Complete mixing occurs when the Gibbs free energy of the mixture is less than the Gibbs free energy of the components. At high temperatures, the polymer and solvent can mix and form a single phase. As the temperature decreases, the solution begins to separate into two phases (Mitchell, 2003).

RESULT AND DISCUSSION

In this paper, the simulation of Gibbs free energy in mixing polymer solution are performed using spreadsheet software. A spreadsheet was chosen to simulate Gibbs free energy because spreadsheets can perform numerical representations in tables. The ability of a spreadsheet to activate and explore numerical, symbolic, and visual, make spreadsheets become an important tool in promoting abstract thinking in students (M. C. Oliveira & Nápoles, 2010). We can obtain a graph of the relationship between the Gibbs free energy of the polymer mixture with the volume fraction of the polymer, and the volume of the solvent more easily using spreadsheet.

The instrument consists of the main spreadsheet and several secondary spreadsheets that interact with the main spreadsheet. We have created two main spreadsheets. The first main spreadsheet shows a Gibbs free energy simulation and the second shows a Gibbs free energy simulation with some interaction parameters. In the secondary spreadsheet, intermediate calculations are performed for the results from the main spreadsheet, source tables are compiled for instrument charts and other charts, apart from those from the main spreadsheet.

The first main spreadsheet shows the results for a set of Gibbs free energy parameters after we have entered certain data. The first main spreadsheet also contains several graphs showing the relationship between the Gibbs free energy, entropy of mixing, and enthalpy of mixing to the volume of polymer fraction and volume of solvent as shown in Figure 1.

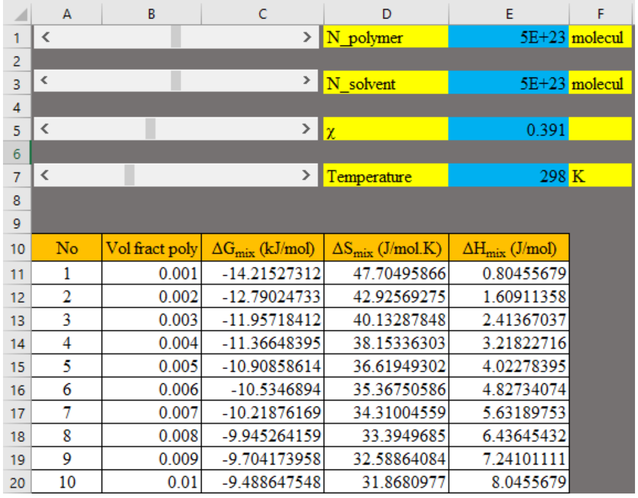


N_polymer	5E+23	molecul
N_solvent	5E+23	molecul
χ	0.391	
Temperature	298	K

No	Vol fract poly	ΔG_{mix} (kJ/mol)	ΔS_{mix} (J/mol.K)	ΔH_{mix} (J/mol)
1	0.001	-14.21527312	47.70495866	0.80455679
2	0.002	-12.79024733	42.92569275	1.60911358
3	0.003	-11.95718412	40.13287848	2.41367037
4	0.004	-11.36648395	38.15336303	3.21822716
5	0.005	-10.90858614	36.61949302	4.02278395
6	0.006	-10.5346894	35.36750586	4.82734074
7	0.007	-10.21876169	34.31004559	5.63189753
8	0.008	-9.945264159	33.3949685	6.43645432
9	0.009	-9.704173958	32.58864084	7.24101111
10	0.01	-9.488647548	31.8680977	8.0455679
11	0.011	-9.293803712	31.21695918	8.85012469
12	0.012	-9.116038344	30.62313096	9.65468148
13	0.013	-8.952614445	30.07742847	10.45923827
14	0.014	-8.801404504	29.57271241	11.26379506
15	0.015	-8.660721971	29.10332323	12.06835185
16	0.016	-8.529207231	28.66469846	12.87290864

Figure 1. First main spreadsheet view

Figure 2 shows the data needed to calculate the Gibbs free energy of mixing polymer solutions. Cells E1, E3, E5, and E7 refer to the number of polymer molecules, the number of solvent molecules, the Fluory-Huggins interaction parameters, and temperature, respectively. These values can be changed using the tools available in the spreadsheet, namely the scroll bar so that the display looks more interactive.



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10	0.01	-9.488647548	31.8680977	8.0455679

Figure 2. Display of input data

Figure 3 shows a graph of the Gibbs free energy against the volume fraction of the polymer. An important function of the change in free energy from one state to another is to determine whether the process is spontaneous or not. When the change in free energy is less than zero ($\Delta G < 0$), the process takes place spontaneously. When the change in free energy is equal to zero ($\Delta G = 0$), the process takes place at equilibrium. When the change in free energy is more than zero ($\Delta G > 0$), the process is not spontaneous (Mitchell, 2003). From Figure 3, the Gibbs free energy has a negative value or less than zero so that the mixing process occurs spontaneously. Complete miscibility occurs when the Gibbs free energy of the mixture is less than the free energy of the components. Figure 4 shows a graphical display of the Gibbs free energy for the number of different polymer molecules.

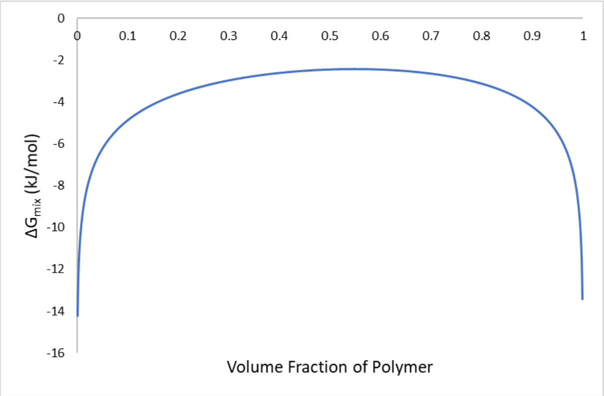
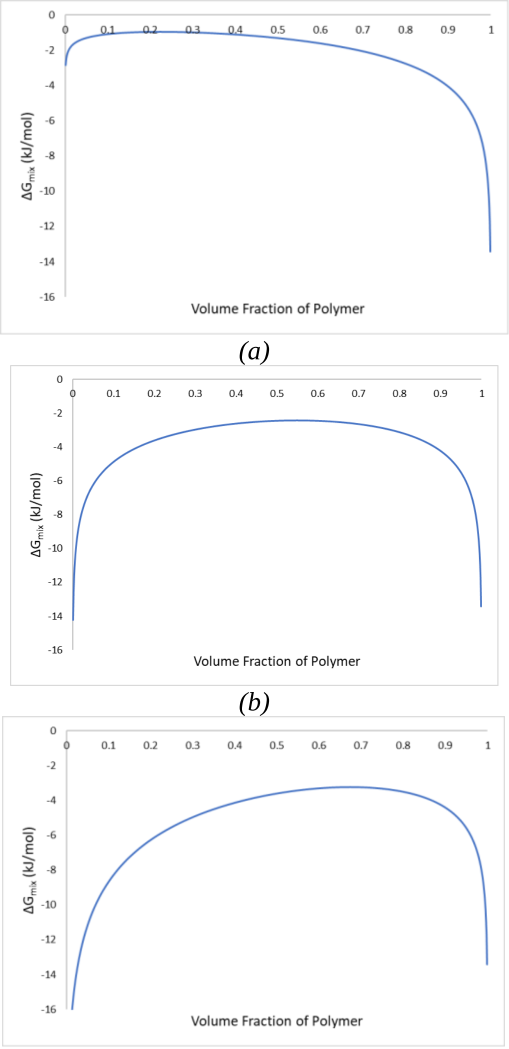


Figure 3. Graph of Gibbs free energy for $N_A = 5 \times 10^{23}$ molecul, $N_B = 10^{23}$ molecul, $\chi = 0,391$, $T = 298$ K

Polymer solutions tend to fall into the category of irregular solutions where ΔS_{mix} and ΔH_{mix} deviate from the ideal. The nonideal behavior can be attributed not only to the presence of a limited heat of mixing but also to the large size difference between the polymer and solvent molecules. The polymer chain can be thought of as a series of covalently bonded small segments, and it is this effect of chain connectivity that causes deviations from the ideal entropy of mixing (Cowie & Arrighi, 2007).



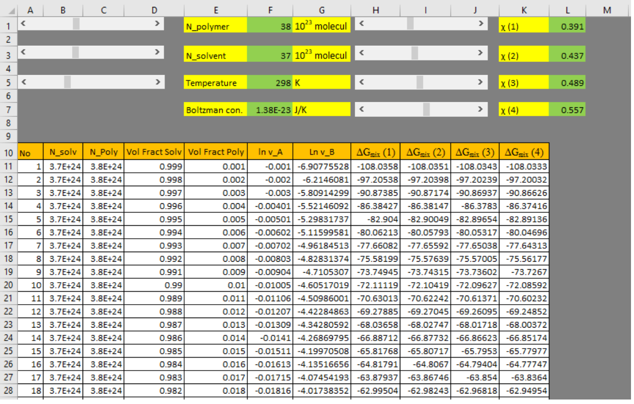
(a)

(b)

(c)

Figure 4. Graph of Gibbs free energy for $N_A = 5 \times 10^{23}$ molecul, $\chi = 0{,}391$, T = 298 K, (a) $N_B = 10^{23}$ molecul, (b) $N_B = 5 \times 10^{23}$ molecul, (c) $N_B = 9 \times 10^{23}$ molecul

Figure 5 shows the second main spreadsheet view. The second main spreadsheet shows the results for a set of Giggs free energy parameters with variations in the value of the Fluory-Huggins interaction parameter. There are four parameters of Fluory-Huggins interaction shown in Figure 3 cells L1, L3, L5, and L7. The interaction parameter values refer to the polymer-solvent interaction parameters shown in Table 1.



N_polymer 38 10^{23} molecul
N_solvent 37 10^{23} molecul
Temperature 298 K
Boltzman con. 1.38E-23 J/K

χ (1) 0.391
χ (2) 0.437
χ (3) 0.489
χ (4) 0.557

No	N_solv	N_Poly	Vol Fract Solv	Vol Fract Poly	ln v_A	Ln v_B	ΔG_{mix} (1)	ΔG_{mix} (2)	ΔG_{mix} (3)	ΔG_{mix} (4)
1	3.7E+24	3.8E+24	0.999	0.001	-0.001	-6.90775528	-108.0358	-108.0351	-108.0343	-108.0333
2	3.7E+24	3.8E+24	0.998	0.002	-0.002	-6.2146081	-97.20538	-97.20398	-97.20239	-97.20032
3	3.7E+24	3.8E+24	0.997	0.003	-0.003	-5.80914299	-90.87385	-90.87174	-90.86937	-90.86626
4	3.7E+24	3.8E+24	0.996	0.004	-0.00401	-5.52146092	-86.38427	-86.38147	-86.3783	-86.37416
5	3.7E+24	3.8E+24	0.995	0.005	-0.00501	-5.29831737	-82.904	-82.90049	-82.89654	-82.89136
6	3.7E+24	3.8E+24	0.994	0.006	-0.00602	-5.11599581	-80.06213	-80.05793	-80.05317	-80.04696
7	3.7E+24	3.8E+24	0.993	0.007	-0.00702	-4.96184513	-77.66082	-77.65592	-77.65038	-77.64313
8	3.7E+24	3.8E+24	0.992	0.008	-0.00803	-4.82831374	-75.58199	-75.57639	-75.57005	-75.56177
9	3.7E+24	3.8E+24	0.991	0.009	-0.00904	-4.7105307	-73.74945	-73.74315	-73.73602	-73.7267
10	3.7E+24	3.8E+24	0.99	0.01	-0.01005	-4.60517019	-72.11119	-72.10419	-72.09627	-72.08592
11	3.7E+24	3.8E+24	0.989	0.011	-0.01106	-4.50986001	-70.63013	-70.62242	-70.61371	-70.60232
12	3.7E+24	3.8E+24	0.988	0.012	-0.01207	-4.42284863	-69.27885	-69.27045	-69.26095	-69.24852
13	3.7E+24	3.8E+24	0.987	0.013	-0.01309	-4.34280592	-68.03658	-68.02747	-68.01718	-68.00372
14	3.7E+24	3.8E+24	0.986	0.014	-0.0141	-4.26869795	-66.88712	-66.87732	-66.86623	-66.85174
15	3.7E+24	3.8E+24	0.985	0.015	-0.01511	-4.19970508	-65.81768	-65.80717	-65.7953	-65.77977
16	3.7E+24	3.8E+24	0.984	0.016	-0.01613	-4.13516656	-64.81791	-64.8067	-64.79404	-64.77747
17	3.7E+24	3.8E+24	0.983	0.017	-0.01715	-4.07454193	-63.87937	-63.86746	-63.854	-63.8364
18	3.7E+24	3.8E+24	0.982	0.018	-0.01816	-4.01738352	-62.99504	-62.98243	-62.96818	-62.94954

Figure 5. Second main spreadsheet view

Figure 6 shows the graph of the relationship between the Gibbs free energy and the volume of the polymer fraction at various χ values. The blue, black, green, and red lines represent the values χ = 0.391, χ = 0.437, χ = 0489, and χ = 0.557. The interaction parameters are an important feature of polymer solution theory. It is an indication of how well the polymer interacts with the solvent. The "bad" solvent has an χ value close to 0.5, while the "good" solvent has a lower, or negative, value. The typical range for most synthetic polymer solutions is 0.3 < χ < 0.6 [17]. The interaction parame is zero for an athermal solution, positive for endothermic mixing, and negative for exothermic mixing (Cowie & Arrighi, 2007).

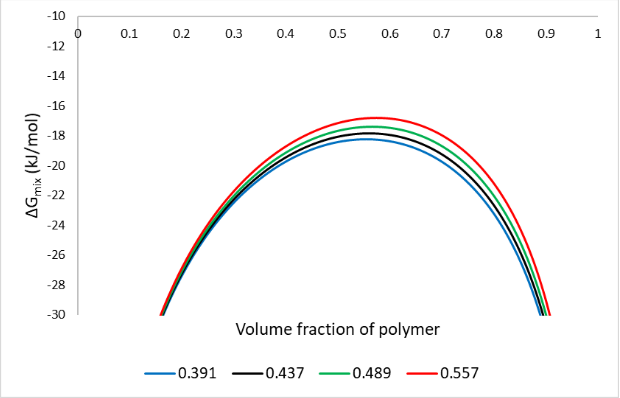


Figure 6. Graph of the relationship of ΔG_{mix} to the volume fraction of polymer at variations in the value of χ

The value of χ can be calculated experimentally for example by using X-rays or neutron scattering and the random phase approximation. Accurate predictions of the χ value can only be obtained from precise liquid-like polymer structures [27]. Table 1 shows some parameters of the polymer-solvent interaction at 25°C.

Table 1. Parameters of polymer-solvent interaction at 25°C (Mitchell, 2003)

Polymer	Solvent	Interaction Parameter χ
cis-Polyisoprene	Toluene (V1 = 106)[a]	0.391
	Benzene (V1 = 89.0)	0.437
Polyisobutylene	Toluene	0.557
	Cyclohexane (V1 = 108)	0.436
Butadiene-styrene 71.5:28.5	Benzene	0.442
	Cyclohexane	0.489
Butadiene-acrylonitrile		
82:18	Benzene	0.390
70:30	Benzene	0.486
61:39	Benzene	0.564

This simple simulation can be used when discussing the topic of polymer thermodynamics. Teachers can start by deriving mathematical equations and then show simulations to visualize the equations. Another option is for students to be asked to create their simulations after deriving a mathematical equation.

Simulation can also be a bridge between theoretical and experimental modeling (Elisabeth Pratidhina et al., 2020).

CONCLUSION

In this article, we show one of the uses of spreadsheets in physics learning, namely simulation of free energy of mixing for a polymer solution. Similar simulations can be done using other applications, but spreadsheets are widely known by teachers and students. Spreadsheets can provide numerical representation in tables so that students can more easily understand equations in the topic of polymer thermodynamics. This simple simulation can be used when discussing the topic of polymer thermodynamics. Teachers can start by deriving mathematical equations and then show simulations to visualize the equations. Another option is for students to be asked to create their simulations after deriving a mathematical equation. Using simulation in learning can make learning more interactiveand help students understand material physics subjects more easily. Spreadsheets can be an alternative for teachers when explaining abstract material to students. Furthermore, simulations with spreadsheets can also support physics learning remotely.

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