**Title: Toward artificial intelligence-based modeling of vapor liquid equilibria of carbon dioxide and refrigerant binary systems**

**Dear Prof. Mirjana Kijevčanin**

**Chemical Engineering Editor**

**Journal of the Serbian Chemical Society**

The authors would like firstly to thank the reviewers for their time and comments. We addressed the comments in the revised version or provided reasoned rebuttals where appropriate. The modified parts/texts have also been highlighted red color to facilitate spotting the changes that have been taken on board.

We are looking forward to hearing from you. Thank you very much for your cooperation.

Sincerely Yours

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**REVIEWER COMMENTS:**

**Reviewer A:**

Does the manuscript contain enough significant original material?

Yes

Is the manuscript clearly and concisely written?

Yes

Are the conclusions adequately supported by the data?

Yes

Does the manuscript give appropriate credit to related recent publications?

Yes

Are the references appropriate and free of important omissions?

Yes

Is the length of the manuscript appropriate?

Yes

Does the manuscript need condensation or extension?

No

Is the quality of the figures (including legends and axes labelling) satisfactory?

Yes

Are the nomenclature and units in accordance with SI?

Yes

Are the English grammar and syntax satisfactory?

Yes

**REVIEWER COMMENT, No. 1:**

Line 277-CONCLUSION

“A MLP neural network with 5-16-1 structure has been used to predict both bubble and dew point pressure of five binary carbon dioxide + refrigerant systems.”

“Therefore, the proposed MLP model can be reliably used to estimate the bubble and dew point pressure of the CO2 + refrigerant systems within the ranges of temperature and pressure considered in this study.” Those two lines are contradictory. Prediction results of dew point pressure for system CO2-R30 are not provided? Those data are not included in training of ANN, and if they are used in testing ANN, results should be presented.

**Response and action:**

Generally, to determine phase diagrams of mixtures, the synthetic method has been used in the past decades (Gonzalez et al., 2002). In this method, a mixture of known composition is prepared in the cell, and its behavior is then observed as a function of temperature and pressure. It should be noted that this method is suitable for bubble points’ determination, since changes of slope are rather sharp. On the other hand, when dew points are considered, changes might be smoother and thus unclear. Therefore, due to this limitation on the experimental observation, the dew points were not reported by Gonzalez et al., 2002. As experimental dew point pressure for system CO2-R30 was not provided, prediction results of this mixture are missing.

In addition, the following sentence:

“A MLP neural network with 5-16-1 structure has been used to predict both bubble and dew point pressure of five binary carbon dioxide + refrigerant systems.”

Is modified as follows:

“A MLP neural network with 5-14-1 structure has been used to predict bubble and dew point pressure of five and four binary carbon dioxide + refrigerant systems, respectively.”

It should be mentioned that for considering comment of the second reviewer the structure of MLP model has changed from 5-16-1 to 5-14-1.

**REVIEWER COMMENT, No. 2:**

line 269-Table III.

Prediction results of dew point pressure for CO2-R30 mixture are missing? Overall DP pressure interval (0.18-9.09), is incorrect according to number of data (239).

**Response and action:**

As mentioned before in the answer to comment No. 1, due to the limitation on the experimental observation, the dew points were not reported by Gonzalez et al., 2002. As experimental dew point pressure for system CO2-R30 was not provided, prediction results of this mixture are missing.

In addition, unfortunately a typo was happened in reporting the overall DP pressure interval. In the revised version of manuscript, the DP pressure interval of 0.18-9.09 is changed to 0.18-7.46. Thank you very much for your comment, please see Table IV in the revised manuscript.

**REPORT:**

This work has only practical significance in modelling five CO2-refrigerant binary systems, and only if authors provide optimised ANN to readers, as supplementary material.

**Response and action:**

For considering your valuable comment, the weights and biases of the optimum ANN structure are tabulated in Table III of the revised version of manuscript. Please see lines 242-246 of the revised manuscript.

In my opinion, this manuscript should:

Be published after major revision and additional review

If manuscript is suitable for publishing, referees recommendation:

Original scientific paper

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**Reviewer B:**

Does the manuscript contain enough significant original material?

Yes

Is the manuscript clearly and concisely written?

Yes

Are the conclusions adequately supported by the data?

Yes

Does the manuscript give appropriate credit to related recent publications?

Yes

Are the references appropriate and free of important omissions?

Yes

Is the length of the manuscript appropriate?

Yes

Does the manuscript need condensation or extension?

Yes

Is the quality of the figures (including legends and axes labelling) satisfactory?

Yes

Are the nomenclature and units in accordance with SI?

Yes

Are the English grammar and syntax satisfactory?

Yes

**ADDITIONAL COMMENTS**

Please indicate the page numbers for suggested corrections. Please, be as specific as possible if major correction by the author(s) is recommended

**REPORT:**

**REVIEWER COMMENT, No. 1:**

I cannot recommend the publication of this paper in its current form. ANNs have emerged as simple and accurate modeling technique, but ANN-based models are valid only if their development were performed with rigorous evaluation. Hence in this paper, the major issue is model testing; as can be seen from Table II so-called test set was actually used for or the adjustment of model parameters i.e. the number of hidden neurons. If you make multiple training runs until you get something that works best on the test data, you have just rendered the test data set as training, and the model you finally create has to be tested once again with NEW test data. In contract, you've essentially created a model specifically for your test set.

Therefore, you must have three datasets: training for weights tuning (e.g. 70% of data), validation for H neurons determination (20% of data) and “unseen” test dataset to evaluate real performance of the final model (use remaining 10% of data).

**Response and action:**

Your comment is considered in the revised version of manuscript. Please see TABLE II and provided explanations in Lines 216-222 and 233-239 in the revised manuscript.

**REVIEWER COMMENT, No. 2:**

Also, it is unclear how many models have been created concerning that “Collected experimental data containing 264 bubble point pressures and 239 dew point pressures…”. I feel that separate models are needed for bubble and dew point pressure?

**Response and action:**

In this study both DP and BP are calculated by a single MLP model.

Input layer of the developed MLP model has five independent variables. Mole fractions of CO2 in liquid and vapor phase are two of these independent variables. It is obvious that yCO2 and xCO2 should be given for DP and BP calculation, respectively. On the other hand, yCO2 in BP calculation, and xCO2 in DP calculation should not be used. Hence, in these circumstances they have been inactivated by setting their values equal to zero. It does not mean that these variables are zero, indeed it has been done to provide a situation for estimation of DP and BP by a single ANN model. Therefore for estimation of DP and BP with a single MLP model,  and has to be zero, respectively.

This explanation is added to the revised version. Please see lines 117-124.

**Minor issues**

**REVIEWER COMMENT, No. 3:**

Abstract: VLE?

**Response and action:**

The VLE is now written out in the Abstract as requested by the reviewer.

**REVIEWER COMMENT, No. 4:**

Line 15. Sensitivity analyses give the significance of inputs, while trial-and-error procedure can be used for the ANN adjustments.

**Response and action:**

"*Sensitivity analyses*" are changed to "*Trial and error procedure*" in the revised version of manuscript.

**REVIEWER COMMENT, No. 5:**

Line 16. “…two-layer neural network…” Two-layer ANN is simple perceptron than can perform only binary classification. A three-layer ANN is MLP.

**Response and action:**

“…two-layer neural network…” is changed to “…three-layer neural network…” in the revised version of the manuscript.

**REVIEWER COMMENT, No. 6:**

Line 48. ANN?

**Response and action:**

The ANN is now written out in the first place is appeared as requested by the reviewer.

**REVIEWER COMMENT, No. 7:**

Fig. 2. should be deleted.

**Response and action:**

Fig. 2 is removed from the revised version of manuscript as suggested by reviewer.

**REVIEWER COMMENT, No. 8:**

Figure 3. should be deleted, since model performance can be determined only using external test set. Training results are not relevant.

**Response and action:**

Fig. 3 is removed from the revised version of manuscript as suggested by reviewer.

**REVIEWER COMMENT, No. 9:**

Figure 5. In fig 5 you must clearly mark the training, val. and test data points. Currently, it is misleading.

**Response and action:**

In Fig. 2 of the revised version of manuscript (Fig. 5 of the old manuscript) training, validation and testing data points are marked as requested by reviewer.

In my opinion, this manuscript should:

Be published after major revision and additional review

If manuscript is suitable for publishing, referees recommendation:

Original scientific paper