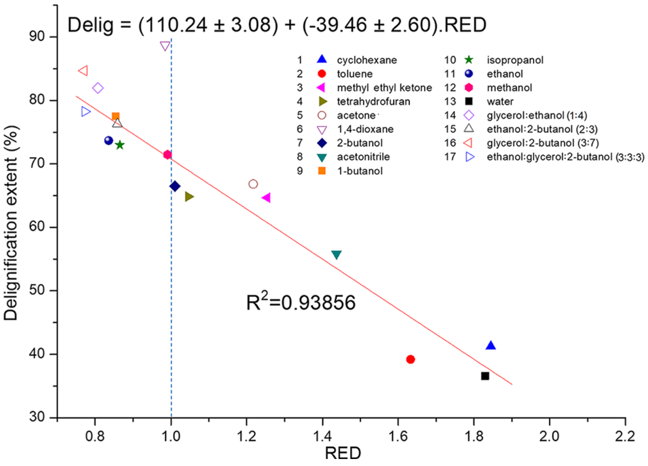

Figure 5. Relative energy difference (RED) for optimized lignin solubility sphere versus delignification extents of organosolv treatments of sugar cane bagasse (linear regression without 1,4-dioxane).

hydrogen bonds. On the other hand, the other organic solvent mixtures that lead to high delignification extents are mainly formed by alcohols with high capacity for hydrogen bonds associated with intermediate values for polar interactions.[23] The mixture with 10% water affects the final properties of the organic solvent:water (9:1) solutions with particular increase of δ_P and δ_H for dioxane:water solutions and minor alterations for alcohol:water solutions.

The optimized HSP values for sugar cane bagasse lignin obtained in this work differs from that one published by Hansen and Björkman.[15] Although both determinations present similar results for dispersive interactions (δ_D) and the sphere radius of solubility (R_0), the polar and hydrogen bond interactions are different in each determination. Even though not completely understood, these differences could be attributed to intrinsic characteristics of sugar cane bagasse lignin. Lignins from sugar cane (as well as lignins from grasses) are classified as HGS lignin and contain higher amount of *p*-hydroxy phenyl moieties, including *p*-coumarates and ferulates.[43] Thus, it is important to assume specific values for HSP in function of the source of lignins.

4. CONCLUSIONS

By adjusting the delignification data to the Hildebrand solubility parameter of the different solvents, it was concluded that this parameter is not the ideal parameter to relate solubility with delignification extents. The Hildebrand parameter does not differentiate the type of interactions that each solvent can perform with lignin. Thus, the Hansen parameters were found as a better way to describe the relationship between the organosolv delignification under the conditions used and the relative energy difference of the center of the lignin solubility sphere to the solvent solubility parameter.

It was also concluded that the Hansen solubility parameters verified in the literature for lignin do not allow a linear fit of the delignification data. Therefore, a new lignin solubility sphere was calculated from delignification yields. With an unoptimized sphere, four solvent mixtures of green and renewable industrial solvents were applied, indicating the possibility of the use of HSP for predicting/choosing solvents for the organossolv delignification of lignocellulosic raw materials. Moreover, these mixtures could also be used to optimize the solubility sphere for the sugar cane bagasse lignin extracted under the specific conditions. The optimized solubility sphere allowed a better data fit with a linear regression determination coefficient of 0.93856, excluding the data from 1,4-dioxane. A specific set of values of HSP for sugar cane bagasse lignin was determined as δ_D = 21.42 MPa$^{1/2}$, δ_P = 8.57 MPa$^{1/2}$, δ_H = 21.80 MPa$^{1/2}$, and R_o = 13.56.

Finally, it was found that because of the difficulties in checking the lignin solubility parameter (it does not exist in native form as an isolated structure), the solubility parameter for the actual lignin cannot be stated with certainty, despite the good fit of the data. However, under the conditions studied, this sphere of solubility allows a better prediction of a delignification extent and the selection of a solvent, considering the desired delignification.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.iecr.9b00875.

Tables S1−S3 (PDF)

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Notes

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