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Buswell's Equation for Quantifying Biohydrogen

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Abstract

Biohydrogen is widely generated by dark fermentation and two-stage anaerobic digestion. Anaerobic digestion can be chemically represented by the Buswell's equation, which is based on elemental composition of organic matter. When compared to Buswell's equation for biomethane, its equation for biohydrogen has been given less attention. In this article, the nature of Buswell's equation for biohydrogen is introduced by using the H-atom method. The mean oxidation number of organic carbons is employed as a metric for counting theoretical quantity of biohydrogen and parameters of Buswell's equation for biohydrogen. Based on an empirical formula and mean oxidation number of organic carbons, the general Buswell's equation for biohydrogen is developed. The mathematical relationships among mean oxidation number of organic carbons, empirical formula, quantity of biohydrogen, theoretical biohydrogen potential, and biohydrogen percent yield are also established. Biowastes and bio-substrates are chosen to demonstrate this notion.

Keywords: Buswell's equation for biohydrogen, H-atom method, mean oxidation number of organic carbons, theoretical biochemical hydrogen potential, biohydrogen percent yield

1. Introduction

Anaerobic digestion is a microbial-mediated process used in waste treatment (Fang, 2010), biofuel production (Kusch-Brandt et al., 2023), and biorefinery chemicals production (Bolzonella et al., 2023; Allaart et al., 2023). As a biofuel, biohydrogen is generated either by microbial-driven biochemical reactions or thermochemical treatment of biomass (Pandey et al., 2013). Dark fermentation (Dzulkarnain et al., 2022; Sarangi & Nanda, 2020) and two-stage anaerobic digestion (Algapani et al., 2019; O-Thong et al, 2018) are widely used for generation of biohydrogen. Anaerobic digestion can be represented by Buswell's equation (BEq), which is based on elemental composition of organic matter (Buswell & Boruff, 1932; Buswell & Mueller, 1952; Boyle, 1977). Although the stoichiometric Buswell's equation is a significant model for counting quantity of biomethane, Buswell's equation for biohydrogen (BEqH) has been relatively less studied (Pererva et al., 2020).

In this article, the H-atom method (Yuen & Lau, 2021) is used to introduce the nature of BEqH through balancing, quantifying, deducting, and defining its stoichiometric equation. The mean oxidation number of organic carbons (ONc) acts as a metric for developing the ONc-BEqH model, in which the mathematical relationships among ONc, empirical formula, quantity of biohydrogen (nH_2), theoretical biohydrogen potential (TBHP), and theoretical biohydrogen yield (TBHY) can be established. The nH_2, nCO_2, ratio of nH_2 to nCO_2, TBHP, and TBHY can also be determined. Biowastes and bio-substrates are chosen to demonstrate the counting of BEqH's parameters, biodegradability index (BDI) and biohydrogen percent yield (BHPY).

2. H-atom Method for Understanding BEqH

The H-atom method can be used to study Buswell's equations (Yuen & Lau, 2023a; 2023b) and to analyze the nature of BEqH.

2.1 Balancing BEqH of Organic Matters

Example 1. Balancing the BEqH for an empirical formula, $C_3H_7O_2NS$

Unbalanced BEqH: $C_3H_7O_2NS+H_2O \rightarrow H_2+CO_2+NH_3+H_2S$

Step 1. Divide overall equation into two half reactions

$$C_3H_7O_2NS \rightarrow CO_2+NH_3+H_2S$$

$$H \rightarrow H_2$$

Step 2. Balance two half reactions (balance C→N→S→O→H atoms)

$C_3H_7O_2NS \rightarrow CO_2+NH_3+H_2S$

$C_3H_7O_2NS \rightarrow 3CO_2+NH_3+H_2S$

$C_3H_7O_2NS+4O \rightarrow 3CO_2+NH_3+H_2S$

$C_3H_7O_2NS+4O \rightarrow 3CO_2+NH_3+H_2S+2H$

$C_3H_7O_2NS+4O+8H \rightarrow 3CO_2+NH_3+H_2S+2H+8H$

$C_3H_7O_2NS+4H_2O \rightarrow 3CO_2+NH_3+H_2S+10H$

$H \rightarrow H_2$

$2H \rightarrow H_2$

Step 3. Combine two half reactions with equivalence of H-atom (LCM = 10)

$(C_3H_7O_2NS+4H_2O \rightarrow 3CO_2+NH_3+H_2S+10H) \times 1$

$(2H \rightarrow H_2) \times 5$

$C_3H_7O_2NS+4H_2O \rightarrow 5H_2+3CO_2+NH_3+H_2S$

2.2 Quantifying BEqH's Parameters: nH_2, nCO_2, $\frac{nH_2}{nCO_2}$, and TBHP

Based on the balanced BEqH of $C_3H_7O_2NS+4H_2O \rightarrow 5H_2+3CO_2+NH_3+H_2S$, the stoichiometric coefficients (SC) and parameters of BEqH can be quantified accordingly.

2.2.1 Identify BEqH's SC

$nH_2 = 5$ and $nCO_2 = 3$ can be identified.

2.2.2 Count the BEqH Ratio (ratio of nH_2 to nCO_2)

The BEqH ratio of nH_2 to nCO_2 ($\frac{nH_2}{nCO_2}$) = $\frac{5}{3}$ can be determined.

2.2.3 Calculate Theoretical Biohydrogen Potential (TBHP)

$$\text{TBHP (at STP, mL/g)} = \frac{22400(nH_2)}{\mu C_3H_7O_2NS}$$

$$\text{TBHP} = \frac{22400(nH_2)}{\mu C_3H_7O_2NS} = \frac{22400(5)}{121.159} = 924.405 \text{ (mL/g)}$$

2.3 Deducting General BEqH of $C_xH_yO_zX_wN_vS_uP_t$

Example 2. Deducting BEqH of $C_xH_yO_zX_wN_vS_uP_t$

Unbalanced BEqH: $C_xH_yO_zX_wN_vS_uP_t+H_2O \rightarrow H_2+CO_2+HX+NH_3+H_2S+H_3PO_4$

Step 1. Divide overall equation into two half reactions

$C_xH_yO_zX_wN_vS_uP_t \rightarrow CO_2+HX+NH_3+H_2S+H_3PO_4$

$2H \rightarrow H_2$

Step 2. Balance two half reactions

$C_xH_yO_zX_wN_vS_uP_t \rightarrow CO_2+HX+NH_3+H_2S+H_3PO_4$

$C_xH_yO_zX_wN_vS_uP_t \rightarrow xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4$

$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)O \rightarrow xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4+(y-w-3v-2u-3t)H$

$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)O+2(2x-z+4t)H \rightarrow xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4+(y-w-3v-2u-3t)H+2(2x-z+4t)H$

$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4+(4x+y-2z-w-3v-2u+5t)H$

$2H \rightarrow H_2$

Step 3. Combine two half reactions with equivalence of H-atom {LCM = 2(4x+y−2z−w−3v−2u+5t)}

$[C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4+(4x+y-2z-w-3v-2u+5t)H] \times 2$

$[2H \rightarrow H_2] \times (4x+y-2z-w-3v-2u+5t)$

$2C_xH_yO_zX_wN_vS_uP_t+2(2x-z+4t)H_2O \rightarrow (4x+y-2z-w-3v-2u+5t)H_2+2xCO_2+2wHX+2vNH_3+2uH_2S+2tH_3PO_4$

Balanced BEqH: $C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow \frac{(4x+y-2z-w-3v-2u+5t)}{2}H_2+xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4$

2.4 Defining Redox Terms of BEqH

By using the H-atom model (IUPAC, 2019), the redox terms of Example 2 are demonstrated in Figure 1.

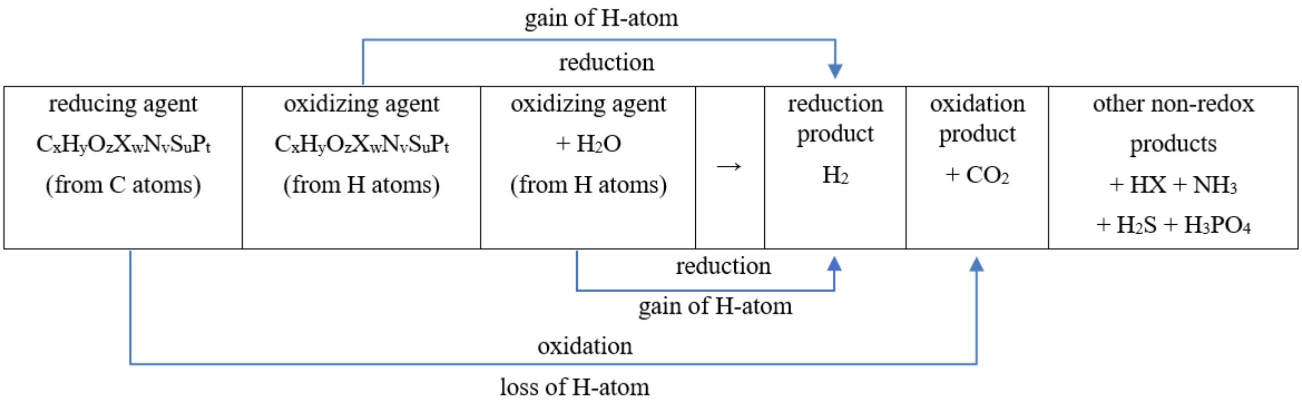


Figure 1. The H-atom method for BEqH

The redox terms of Example 2 are summarized in Table 1. $C_xH_yO_zX_wN_vS_uP_t$ molecules not only gain H-atom (from H atoms) to form hydrogen gas, but also lose H-atom (from C atoms) to produce carbon dioxide. In addition, the H-atom of H_2O gains one H-atom from $C_xH_yO_zX_wN_vS_uP_t$ to form hydrogen gas. There are three sets of redox couples in BEqH.

Table 1. The redox terms of the Buswell's equation for biohydrogen

Redox term	Chemical language	Chemical meaning
Oxidation	$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4+(4x+y-2z-w-3v-2u+5t)H$	loss of H-atom
Reduction	$C_xH_yO_zX_wN_vS_uP_t$ (H-atom)+H→H_2	gain of H-atom
Reduction	H_2O (H-atom)+H→H_2	gain of H-atom
Reducing agent	$C_xH_yO_zX_wN_vS_uP_t$ (C-atom)	loss of H-atom
Oxidizing agent	$C_xH_yO_zX_wN_vS_uP_t$ (H-atom)	gain of H-atom
Oxidizing agent	H_2O (H-atom)	gain of H-atom
Redox reaction	$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow \frac{(4x+y-2z-w-3v-2u+5t)}{2}H_2+xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4$	oxidation + reduction + reduction

2.5 Nature of BEqH

BEqH is an overall microbial-mediated biochemical redox reaction. It is represented in the form of a stoichiometric molecular chemical equation. BEqH is composed of three sets of redox couples. All non-carbon and non-hydrogen atoms are non-redox atoms. Water is involved in the reaction. In the absence of O_2, carbon atoms lose H atoms to form CO_2 and hydrogen atoms gain H atoms to form H_2. H_2O molecules also gain H atoms from organic matter to form H_2 molecules.

$$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow \frac{(4x+y-2z-w-3v-2u+5t)}{2}H_2+xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4$$

Assumptions of the model:

- BEqH is a redox reaction for pure or mixed organic matters in the form of an empirical formula.
- Organic carbons are fully converted to CO_2.
- All halogen, nitrogen, sulfur, and phosphorous (X, N, S and P) atoms are converted to HX, NH_3, H_2S and H_3PO_4, respectively.

3. ONc-BEqH Model

The ONc-BEq model for biomethane was established by integrating the concept of ONc and BEq model (Yuen & Lau, 2024a; 2024b). The same method is applied to construct the analogous ONc-BEqH model. The mathematical equations for the parameters of BEqH are shown below.

3.1 ONc of Empirical Formula, $C_xH_yO_zX_wN_vS_uP_t$

The ONc can be determined by using the non-carbon atom method (Yuen & Lau, 2023b). Regarding empirical formulas $C_xH_yO_zX_wN_vS_uP_t$ of organic matters, individual oxidation number of hydrogen, oxygen, halogen, nitrogen, sulfur, and phosphorous (H, O, X, N, S, and P) atoms are assumed to be +1, −2, −1, −3, −2 and +5 respectively.

$$\because \text{ONc} = \frac{-\Sigma \text{ON}_{\text{inc}}}{\text{nc}} \text{ and nc} = x$$

$$\therefore x\ \text{ONc}(C_xH_yO_zX_wN_vS_uP_t) = -(y-2z-w-3u-2v+5t)$$

$$\text{ONc} = \frac{-(y-2z-w-3u-2u+5t)}{x}$$

3.2 BEqH and ONc-BEqH Models: Stoichiometric Chemical Equations

In the conversion of BEqH to ONc-BEqH, their stoichiometric chemical equations are shown as follows:

$$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow \frac{(4x+y-2z-w-3v-2u+5t)}{2}H_2+xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4$$

$$C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow \frac{x(4-\text{ONc})}{2}H_2+xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4$$

3.3 Parameters of ONc-BEq: nH_2, nCO_2, and $\frac{nH_2}{nCO_2}$

$$nCO_2 = nc = x$$

$$nH_2 = \frac{4x+y-2z-w-3v-2u+5t}{2} = \frac{x(4-\text{ONc})}{2}$$

$$\frac{nH_2}{nCO_2} = \frac{(4-\text{ONc})}{2}$$

3.4 Theoretical Biohydrogen Potential (TBHP)

The theoretical biohydrogen potential (TBHP) for nH_2 in BEqH is analogous to theoretical biochemical methane potential (TBMP) for nCH_4 in BEq (Angelidaki & Sanders, 2004).

$$\text{TBHP (at STP, mL/g)} = \frac{22400(nH_2)}{\mu C_xH_yO_zX_wN_vS_uP_t}$$

$$\text{TBHP (at STP, mL/g)} = \frac{22400\,x(4-\text{ONc})}{2\mu C_xH_yO_zX_wN_vS_uP_t} = \frac{11200\,x(4-\text{ONc})}{\mu C_xH_yO_zX_wN_vS_uP_t}$$

4. From Counting ONc to Determining nH_2, $\frac{nH_2}{nCO_2}$, and TBHP

In Figure 2, the working scheme shows how empirical formula is converted to nH_2, $\frac{nH_2}{nCO_2}$, and TBHP. Example 3 demonstrates the calculation.

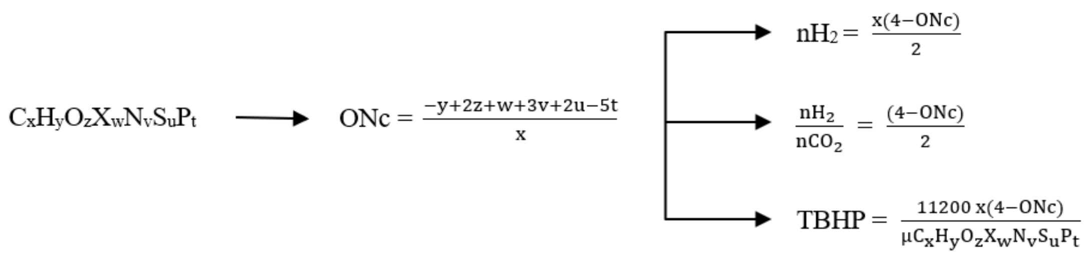


Figure 2. Working scheme: empirical formula → ONc → BEqH's parameters

Example 3. Determine nH_2, $\frac{nH_2}{nCO_2}$, and TBHP of a given empirical formula, $C_3H_7O_2NS$

Solve: (i) by using equation: ONc ($C_xH_yO_zX_wN_vS_uP_t$) = $\frac{-y+2z+w+3v+2u-5t}{x}$

For $C_xH_yO_zN_vS_u = C_3H_7O_2NS$

Atom	C	H	O	N	S
ON	-	+1	-2	-3	-2
AC	3	7	2	1	1

$$ONc\ (C_3H_7O_2NS) = \frac{-y+2z+3v+2u}{x} = \frac{-[(7)-2(2)-3(1)-2(1)]}{3} = +\frac{2}{3}$$

(ii) by using equation: $nH_2 = \frac{x(4-ONc)}{2}$

$ONc = +\frac{2}{3}$, $x = 3$

$$nH_2\ (mol) = \frac{(3)(4-(+\frac{2}{3}))}{2} = 5$$

(iii) by using equation: $\frac{nH_2}{nCO_2} = \frac{(4-ONc)}{2}$

$ONc = +\frac{2}{3}$

$$\frac{nH_2}{nCO_2} = \frac{(4-(+\frac{2}{3}))}{2} = \frac{5}{3}$$

(iv) by using equation: $TBHP = \frac{11200\,x(4-ONc)}{\mu}$

$\mu C_3H_7O_2NS$ (g/mol) = 121.159 and x = 3

$$TBHP\ (mL/g\ at\ STP) = \frac{11200\ (3)(4-(+\frac{2}{3}))}{(121.159)} = 924.405$$

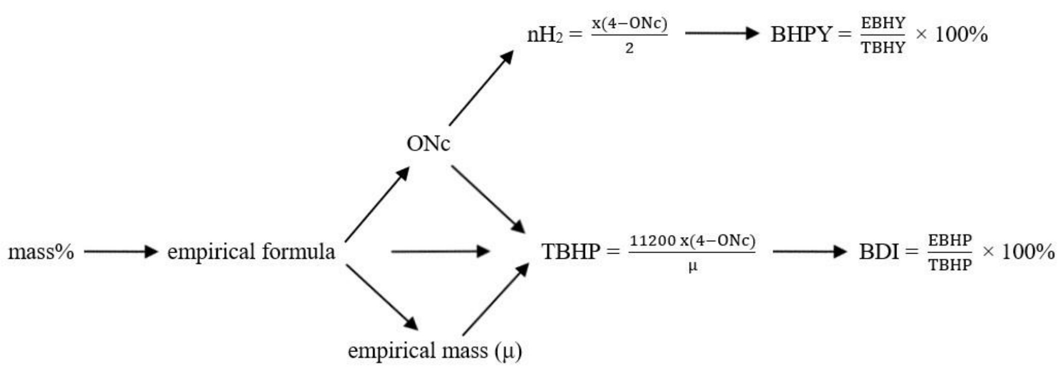
5. Applications of ONc-BEqH

5.1 Operating Scheme

When the mass% of elements are identified, organic matter's empirical formula and ONc can be determined. Consequently, the empirical formula mass (μ), nH_2 and theoretical biohydrogen potential (TBHP) can be counted. Figure 3 shows the operating scheme. The biodegradability index for nH_2 in BEqH is analogous to biodegradability index for nCH_4 in BEq (Nielfa et al., 2015). The biodegradability index for biohydrogen (BDI) and biohydrogen percent yield (BHPY) can then be calculated.

$BDI = \frac{EBHP}{TBHP} \times 100\%$	$BHPY = \frac{EBHY}{TBHY} \times 100\%$
EBHP = experimental biohydrogen potential	EBHY = experimental biohydrogen yield = experimental nH_2
TBHP = theoretical biohydrogen potential	TBHY = theoretical biohydrogen yield = theoretical nH_2

Figure 3. Relationships among mass%, empirical formula, empirical formula mass (μ), ONc, nH_2, TBHP, BDI, and BHYP



5.2 Data Collection

The C/H/O/X/N/S/P contents of most organic matters are attained by elemental analysis. The selected ultimate analysis of dried biowastes which include the six elements of C/H/O/Cl/N/S (Achinas & Euverink, 2016) and C/H/O/N/S/P (Zaher et al., 2010) are retrieved. The empirical formulas ($C_xH_yO_zCl_wN_vS_uP_t$) of selected biowastes are calculated from mass% of elements (Yuen & Lau, 2023b; 2024a). The data are listed in Table 2.

Table 2. Selected biowastes: ultimate analysis and their calculated empirical formula

Biowaste	Mass%									Empirical formula
	Total%	Ash%	C%	H%	O%(*)	Cl%	N%	S%	P%	$C_xH_yO_zCl_wN_vS_uP_t$
Chicken litter	100	15.49	45.32	5.85	27.38	0.35	5.16	0.45	-	$C_{3.773}H_{5.804}O_{1.711}Cl_{0.010}N_{0.368}S_{0.014}$
Feedlot manure	100	15.87	45.39	5.35	30.98	1.16	0.96	0.29	-	$C_{3.779}H_{5.308}O_{1.936}Cl_{0.033}N_{0.069}S_{0.009}$
Milk Cow Manure	100	8.42	44.70	5.90	37.96	-	2.24	0.30	0.48	$C_{3.722}H_{5.853}O_{2.373}N_{0.160}S_{0.009}P_{0.015}$
Horse Manure	100	17.78	46.90	4.20	28.20	-	1.20	1.50	0.22	$C_{3.905}H_{4.167}O_{1.763}N_{0.086}S_{0.047}P_{0.007}$
Beef Cow Manure	100	14.90	45.40	5.40	30.97	-	2.56	0.29	0.48	$C_{3.780}H_{5.357}O_{1.936}N_{0.183}S_{0.009}P_{0.015}$
Poultry Manure	100	13.02	39.57	5.11	35.20	-	2.93	0.77	3.40	$C_{3.294}H_{5.069}O_{2.200}N_{0.209}S_{0.024}P_{0.110}$

Remark: * O% are recalculated by the mathematical equation, O% = 100% − ash% − (C%+H%+Cl%+N%+S%+P%)

5.3 Calculation of ONc and BEqH Parameters

With reference to Example 3, the ONc, nH_2, TBHP, and $\frac{nH_2}{nCO_2}$ of selected biowastes can be calculated and are shown in Table 3.

Table 3. Selected biowastes: calculated ONc, nH_2, TBHP, and $\frac{nH_2}{nCO_2}$

Biowaste	nC = x	$\mu C_xH_yO_zCl_wN_vS_uP_t$	ONc	nH_2	TBHP	$\frac{nH_2}{nCO_2}$
Chicken litter	3.773	84.500	−0.329	8.166	2164.709	2.164
Feedlot manure	3.779	84.139	−0.312	8.147	2168.941	2.156
Milk Cow Manure	3.722	91.565	−0.184	7.786	1904.734	2.092
Horse Manure	3.905	82.238	−0.083	7.972	2171.415	2.041
Beef Cow Manure	3.780	85.092	−0.263	8.057	2120.830	2.131
Poultry Manure	3.294	86.976	−0.165	6.860	1766.745	2.083

6. Biodegradability Index for Biohydrogen (BDI) and Biohydrogen Percent Yield (BHPY)

6.1 Data Collection

The experimental biohydrogen potential (EBHP) and experimental biohydrogen yield (EBHY) of bio-substrates are retrieved from literature (Sarangi & Nanda, 2020).

6.2 Calculation of Biodegradability Index for Biohydrogen (BDI)

$$\text{BDI} = \frac{\text{EBHP (Experimental biohydrogen potential)}}{\text{TBHP (Theoretical biohydogen potential)}} \times 100\%$$

When the experimental biohydrogen potential (EBHP) of bio-substrate is identified, the BDI can be counted. Three sets of calculated $\mu C_xH_yO_z$, ONc, nH_2, theoretical biohydrogen potential (TBHP), and BDI are summarized in Table 4.

Table 4. Selected bio-substrates for molecular formula $C_xH_yO_z$: calculated $\mu C_xH_yO_z$, ONc, nH_2, TBHP, and BDI

Bio-substrate	Molecular formula	$\mu C_xH_yO_z$	ONc	nH_2	TBHP = Theoretical biohydrogen potential (mL/g)	* EBHP = Experimental biohydrogen potential (mL/g)	BDI = $\frac{EBHP}{TBHP}$ × 100%
glycerol	$C_3H_8O_3$	92.094	−0.667	7.000	1702.608	172.9	10.16%
glucose	$C_6H_{12}O_6$	180.156	0.000	12.000	1492.040	124.5	8.344%
xylose	$C_5H_{10}O_5$	150.130	0.000	10.000	1492.040	117.9	7.902%

6.3 Calculation of Biohydrogen Percent Yield (BHPY)

$$\text{BHPY} = \frac{\text{EBHY (Experimrntal biohydrogen yield)}}{\text{TBHY (Theoretical biohydrogen yield)}} \times 100\% = \frac{\text{EBHY (Experimetal } nH_2)}{\text{TBHY (Theoretical } nH_2)} \times 100\%$$

When the experimental biohydrogen yield (EBHY) of bio-substrate is identified, the BHBY can be counted. The value of EBHY is equal to nH_2. Two sets of calculated $\mu C_xH_yO_z$, ONc, theoretical biohydrogen yield (TBHY), and BHPY are summarized in Table 5.

Table 5. Selected bio-substrates for molecular formula $C_xH_yO_z$: calculated $\mu C_xH_yO_z$, ONc, TBHY, and BHPY (%)

Bio-substrate	Molecular formula	$\mu C_xH_yO_z$	ONc	nH_2 = TBHY = Theoretical biohydrogen yield, (mol/mol)	* EBHY = Experimental biohydrogen yield, (mol/mol)	BHPY = $\frac{EBHY}{TBHY}$ × 100%
lactose	$C_{12}H_{22}O_{11}$	342.297	0.000	24.000	1.50	6.25%
cellulose	$C_6H_{10}O_5$	162.141	0.000	12.000	1.36	11.3%

7. Conclusion

The nature of Buswell's equation for biohydrogen (BEqH) is explored by using the H-atom method. The general ONc-BEqH is established as $C_xH_yO_zX_wN_vS_uP_t+(2x-z+4t)H_2O \rightarrow \frac{x(4-ONc)}{2}H_2+xCO_2+wHX+vNH_3+uH_2S+tH_3PO_4$. For any given empirical formula of an organic matter, its BEqH parameters can be calculated by the following mathematical equations: $nH_2 = \frac{(4x+y-2z-w-3v-2u+5t)}{2} = \frac{x(4-ONc)}{2}$, $\frac{nH_2}{nCO_2} = \frac{(4-ONc)}{2}$, and TBHP $= \frac{22400(nH_2)}{\mu C_xH_yO_zX_wN_vS_uP_t} = \frac{11200\,x(4-ONc)}{\mu C_xH_yO_zX_wN_vS_uP_t}$. In addition, the biohydrogen percent yield is defined as BHPY $= \frac{\text{Experimetal biohydrogen yield}}{\text{Theoretical biohydrogen yield}} \times 100\%$ and the biodegradability index as BDI $= \frac{\text{Experimental biohydrogen potential}}{\text{Theoretical biohydogen potential}} \times 100\%$.

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Authors contributions

Dr. Pong Kau Yuen was responsible for designing the study, drafting, and revising the manuscript. Dr. Cheng Man Diana Lau was responsible for revising the manuscript. All authors read and approved the final manuscript.

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Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Informed consent

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Data availability statement

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

Data sharing statement

No additional data are available.

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