
Preface	3
Summary	5
Notation and Nomenclature	19
1 Introduction	21
1.1 Rationale	21
1.2 Contributions of the Thesis	22
1.3 Thesis Outline	23
1.3.1 Part I: Design	23
1.3.2 Part II: Operation	24
Part I: Design	25
2 Distillation Theory	27
2.1 Introduction	28
2.2 Fundamentals	29
2.2.1 The Equilibrium Stage Concept	29
2.2.2 Vapour-Liquid Equilibrium (VLE)	29
2.2.3 K-values and Relative Volatility	31
2.2.4 Estimating the Relative Volatility From Boiling Point Data	32
2.2.5 Material Balance on a Distillation Stage	34
2.2.6 Assumption about Constant Molar Flows	36
2.3 The Continuous Distillation Column	36
2.3.1 Degrees of Freedom in Operation of a Distillation Column	37
2.3.2 External and Internal Flows	38
2.3.3 McCabe-Thiele Diagram	38
2.3.4 Typical Column Profiles — Not optimal feed location	40
2.4 Simple Design Equations	41
2.4.1 Minimum Number of Stages — Infinite Energy	41
2.4.2 Minimum Energy Usage — Infinite Number of Stages	42
2.4.3 Finite Number of Stages and Finite Reflux	43
2.4.4 Constant K-values — Kremser Formulas	44

2.4.5	Approximate Formula with Constant Relative Volatility . . .	45
2.4.6	Optimal Feed Location	47
2.4.7	Summary for Continuous Binary Columns	48
2.5	Multicomponent Distillation — Underwood’s Method	51
2.5.1	The Basic Underwood Equations	51
2.5.2	Stage to Stage Calculations	53
2.5.3	Some Properties of the Underwood Roots	54
2.5.4	Minimum Energy — Infinite Number of Stages	55
2.6	Further Discussion of Specific Issues	58
2.6.1	The Energy Balance and Constant Molar Flows	58
2.6.2	Calculating Temperature when Using Relative Volatilities .	60
2.6.3	Discussion and Caution	62
2.7	Bibliography	62
3	Analytic Expressions and Visualization of Minimum Energy Consumption in Multicomponent Distillation: A Revisit of the Underwood Equations.	63
3.1	Introduction	64
3.1.1	Background	64
3.1.2	Problem Definition - Degrees of Freedom	65
3.2	The Underwood Equations for Minimum Energy	65
3.2.1	Some Basic Definitions	65
3.2.2	Definition of Underwood Roots	66
3.2.3	The Underwood Roots for Minimum Vapour Flow	67
3.2.4	Computation Procedure	68
3.2.5	Summary on Use of Underwood’s Equations	72
3.3	The V_{min} -diagram (Minimum Energy Mountain)	73
3.3.1	Feasible Flow Rates in Distillation	74
3.3.2	Computation Procedure for the Multicomponent Case	75
3.3.3	Binary Case	75
3.3.4	Ternary Case	78

3.3.5	Five Component Example	81
3.3.6	Simple Expression for the Regions Under the Peaks	82
3.4	Discussion	83
3.4.1	Specification of Recovery vs. Composition	83
3.4.2	Behaviour of the Underwood Roots	83
3.4.3	Composition Profiles and Pinch Zones	85
3.4.4	Constant Pinch-zone Compositions (Ternary Case)	85
3.4.5	Invariant Multicomponent Pinch-zone Compositions	89
3.4.6	Pinch Zones for $V > V_{\min}$	90
3.4.7	Finite Number of Stages	90
3.4.8	Impurity Composition with Finite Number of Stages	92
3.5	Summary	92
3.6	References	93
4	Minimum Energy for Three-product Petlyuk Arrangements	95
4.1	Introduction	96
4.2	Background	97
4.2.1	Brief Description of the Underwood Equations	97
4.2.2	Relation to Previous Minimum Energy Results	98
4.2.3	The V_{\min} -diagram for Conventional Columns	99
4.3	The Underwood Equations Applied to Directly Coupled Sections	100
4.3.1	The Petlyuk Column Prefractionator	100
4.3.2	Composition Profiles	101
4.3.3	Reverse Net Flow of Components	102
4.3.4	Reverse Flow Effects on the Underwood Roots	104
4.4	“Carry Over” Underwood Roots in Directly Coupled Columns	105
4.5	V_{\min} -Diagram for Directly Coupled Columns	108
4.6	Minimum Energy of a Ternary Petlyuk Arrangement	110
4.6.1	Coupling Column C22 with Columns C21 and C1	110
4.6.2	Visualization in the V_{\min} -diagram	112

4.6.3	Nonsharp Product Specifications	115
4.6.4	The Flat Optimality Region	115
4.7	Improved 2nd Law Results in Petlyuk Arrangements	117
4.8	Minimum Energy with Multicomponent Feed	118
4.8.1	The General Rule	119
4.8.2	Example: Sharp Component Splits in Products	119
4.8.3	Example: Nonsharp Product Split	121
4.9	Discussion	122
4.9.1	The Conventional Reference	122
4.9.2	Extra Condenser or Reboiler in the Prefractionator	123
4.9.3	Use of a Conventional Prefractionator Column	125
4.9.4	Heat Integration	125
4.9.5	The Two-Shell Agrawal Arrangement	126
4.9.6	A Simple Stage Design Procedure	126
4.9.7	Possible Reduction of Stages	127
4.9.8	Short Note on Operation and Control	129
4.10	Conclusion	130
4.11	References	131
5	Minimum Energy for Separation of Multicomponent Mixtures in Directly Coupled Distillation Arrangements	135
5.1	Introduction	136
5.2	Four Components and Four Products	137
5.2.1	Extended Petlyuk Arrangement	137
5.2.2	Minimum Vapour Flow Expressions	138
5.2.3	Visualization in the V_{min} -Diagram	140
5.2.4	The Highest Peak Determines the Minimum Vapour Flow	142
5.2.5	Composition at the Junction C21-C22-C32	143
5.2.6	Flows at the Feed Junction to C32	144
5.2.7	Composition Profile - Simulation Example	145
5.3	Minimum Energy for N Components and M Products	146

5.3.1	Vmin for N Feed Components and N Pure Products	147
5.3.2	General Vmin for N Feed Components and M Products . . .	148
5.4	Verification of the Minimum Energy Solution	150
5.4.1	Minimum Vapour Flow as an Optimization Problem	151
5.4.2	Requirement for Feasibility	151
5.4.3	Verification of The Optimal Solution	152
5.4.4	Summary of the Verification	155
5.4.5	The Optimality Region	156
5.5	Discussion	157
5.5.1	Arrangement Without Internal Mixing	157
5.5.2	Practical Petlyuk Arrangements (4-product DWC).	159
5.5.3	Heat Exchangers at the Sidestream Junctions	162
5.5.4	The Kaibel column or the “ column”	163
5.5.5	Required Number of Stages - Simple Design Rule	163
5.5.6	Control	164
5.6	Conclusion	164
5.7	References	165
6	Minimum Energy Consumption in Multicomponent Distillation	169
6.1	Introduction	170
6.1.1	Some Terms	170
6.1.2	Basic Assumptions	171
6.1.3	Minimum Entropy Production (2nd law efficiency)	172
6.1.4	Minimum Energy (1st law)	173
6.1.5	Summary of some Computation Examples	174
6.2	The Best Adiabatic Arrangement Without Internal Heat Exchange .	176
6.2.1	Direct Coupling Gives Minimum Vapour Flow	176
6.2.2	Implications for Side-Strippers and Side-Rectifiers	179
6.2.3	The Adiabatic Petlyuk Arrangement is Optimal	179
6.3	Entropy Production in Adiabatic Arrangements	180

6.3.1	Adiabatic Column (Section)	180
6.3.2	Adiabatic Petlyuk Arrangements	181
6.4	Reversible Distillation	182
6.4.1	The Reversible Petlyuk Arrangement	183
6.4.2	Comparing Reversible and Adiabatic Arrangements	187
6.5	A Case Study: Petlyuk Arrangements with Internal Heat Exchange	188
6.5.1	Example 0: Theoretical Minimum Energy Limit	188
6.5.2	Example 1: Internal Heat Exchange in the Reversible Arrangement	188
6.5.3	Example 2: Heat Exchange Across the Dividing Wall	189
6.5.4	Example 3: Pre-heating of the Feed by Heat Exchange with the Sidestream	190
6.5.5	Summary of The Examples	191
6.6	Operation at Several Pressure Levels	192
6.6.1	Example 1: Feed Split (Binary Case)	192
6.6.2	Example 2: Double Effect Direct Split (DEDS)	193
6.6.3	Example 3: Double Effect Prefractionator Column (DEPC)	194
6.6.4	Relation to the Petlyuk Column and the V_{min} -diagram	194
6.7	Discussion	196
6.7.1	Plant-wide Issues	196
6.7.2	Heat Exchange at the Sidestream Stages	196
6.7.3	Non-Uniqueness of Heat Supply in Reversible Columns	197
6.7.4	Practical Issues	199
6.8	Conclusion	199
6.9	References	200
6.10	Appendix: Reversible Distillation Theory	201
6.10.1	Temperature-Composition-Pressure Relationship	202
6.10.2	The Reversible Vapour Flow Profile	203
6.10.3	Entropy Production in a Reversible Section	204
6.10.4	Reversible Binary Distillation	205

Part II: Operation **209**

7 Optimal Operation of Petlyuk Distillation: Steady-State Behaviour	211
7.1 Introduction	212
7.2 The Petlyuk Column Model	215
7.3 Optimization Criterion	216
7.3.1 Criterion with State Space Model	217
7.4 Results From the Model Case Study	218
7.4.1 Optimal Steady State Profiles	218
7.4.2 The Solution Surface	220
7.4.3 Effect of Disturbances	222
7.4.4 Transport of Components	222
7.5 Analysis from Model with Infinite Number of Stages	224
7.5.1 Minimum Energy Consumption for a Petlyuk Column. ...	225
7.5.2 Solution Surface for Infinite Number of Stages	226
7.5.3 Analyzing the Effect of the Feed Enthalpy	230
7.5.4 How Many Degrees of Freedom Must we Adjust During Operation?	230
7.5.5 Sensitivity to Disturbances and Model Parameters	233
7.5.6 A Simple Control Strategy with one Degree of Freedom Fixed	233
7.5.7 Liquid Fraction: Bad Disturbance or Extra Degree of Freedom?	234
7.5.8 Relations to Composition Profiles	234
7.6 Candidate Feedback Variables	236
7.6.1 Position of Profile in Main Column (Y1).	236
7.6.2 Temperature Profile Symmetry (Y2)	237
7.6.3 Impurity of Prefractionator Output Flows (Y3,Y4)	238
7.6.4 Prefractionator Flow Split (Y5)	238
7.6.5 Temperature Difference over Prefractionator (Y6)	241
7.6.6 Evaluation Of Feedback Candidates	243

7.7	Conclusions	243
7.8	Acknowledgements	243
7.9	References	243
7.10	Appendix	244
7.10.1	Model Equations for the Finite Dynamic Model	244
7.10.2	Analytic Expressions for Minimum Reflux	246
7.10.3	Mapping $V(b,L1)$ to $V(Rl,Rv)$	249
8	Use of Short-cut Methods to Analyse Optimal Operation of Petlyuk Distillation Columns	251
8.1	Introduction	252
8.2	The Petlyuk Distillation Column	252
8.3	Computations with Infinite Number of Stages	253
8.4	Results with the Analytical Methods or some Separation Cases	256
8.4.1	When do we get the Largest Savings with the Petlyuk Column?	256
8.4.2	Sensitivity to Changes in Relative Volatility Ratio and Liquid Fraction	258
8.4.3	When Can we Obtain Full Savings with Constant Vapour and Liquid Splits?	258
8.5	A Simple Procedure to Test the Applicability for a Petlyuk Arrangement	260
8.6	CONCLUSION	261
8.7	ACNOWLEDGEMENT	261
8.8	REFERENCES	261
9	Optimal Operating Regions for the Petlyuk Column - Nonsharp Specifications	263
9.1	Introduction	264
9.2	The Basic Methods	265
9.2.1	The Underwood Equations	265
9.2.2	The V_{min} -Diagram	266
9.2.3	The V_{min} -diagram Applied to the Petlyuk Arrangement	266

9.2.4	The Optimality Region for Sharp Product Splits	267
9.3	Non-Sharp Product Specifications	268
9.3.1	Relation Between Compositions, Flows and Recoveries . . .	268
9.4	Minimum Vapour Flow for Non-Sharp Product Specifications . . .	269
9.5	The Optimality Region	272
9.5.1	Possible Impurity Paths to the Sidestream	272
9.5.2	The Optimality Region for Case 1	273
9.5.3	Net Flow of Heavy C into Top of Column C22	275
9.5.4	Optimality Regions for Case 3	276
9.5.5	Optimality region for Case 2 (Balanced Main Column) . . .	277
9.5.6	Effect of the Feed Composition	277
9.5.7	Sensitivity to Impurity Specification-Example	278
9.6	Operation Outside the Optimality Region	278
9.6.1	The Solution Surface - Simulation Example	279
9.6.2	Characteristics of the Solution	280
9.6.3	Four Composition Specifications	281
9.6.4	Failure to Meet Purity Specifications	283
9.7	Conclusions	284
9.8	References	284
9.9	Appendix: Alternative Proof of the Optimality Region for Case 1	285
10	Self-Optimizing Control:	
	Local Taylor Series Analysis	287
10.1	Introduction	288
10.1.1	The Basic Idea	288
10.2	Selecting Controlled Variables for Optimal Operation	289
10.2.1	The Performance Index (cost) J	289
10.2.2	Open-loop Implementation	291
10.2.3	Closed-loop Implementation	292
10.2.4	A Procedure for Output Selection (Method 1)	294

10.3 Local Taylor Series Analysis	296
10.3.1 Expansion of the Cost Function	296
10.3.2 The Optimal Input	298
10.3.3 Expansion of the Loss Function	299
10.3.4 Loss With Constant Inputs	299
10.3.5 Loss with Constant Controlled Outputs	300
10.3.6 Loss Formulation in Terms of Controlled Outputs	301
10.3.7 “Ideal” Choice of Controlled Outputs	302
10.4 A Taylor-series Procedure for Output Selection	303
10.5 Visualization in the Input Space	305
10.6 Relationship to Indirect and Partial Control	307
10.7 Maximizing the Minimum Singular Value (Method 2)	310
10.7.1 Directions in the Input Space	311
10.7.2 Analysis in the Output Space	312
10.8 Application Examples	313
10.8.1 Toy Example	313
10.8.2 Application to a Petlyuk Distillation Column	314
10.9 Discussion	316
10.9.1 Trade-off in Taylor Series Analysis	316
10.9.2 Evaluation of Loss	316
10.9.3 Criterion Formulation with Explicit Model Equations	317
10.9.4 Active Constraint Control	318
10.9.5 Controllability Issues	319
10.9.6 Why Separate into Optimization and Control	319
10.10References	320
11 Evaluation of self-optimising control structures for an integrated Petlyuk distillation column	323
11.1 Introduction	324
11.2 Energy Optimization in the Petlyuk Column	324
11.3 Optimising Control Requirement for the Petlyuk Column	325

11.4 Self-optimising Control for the Petlyuk Column	326
11.5 Self-optimising Control: A Petlyuk Column Case Study	327
11.5.1 The Nominal Optimal Solution	327
11.5.2 Proposed Output Feedback Variables	328
11.6 Robustness Study Simulation	329
11.7 Discussion of the Results	330
11.8 Conclusions	331
11.9 References	331
12 Conclusions and Further Work	335
12.1 Contributions	335
12.2 Further Work	337
12.2.1 Process Design	337
12.2.2 Control Structure Design	337
12.2.3 Advanced Control	338
12.3 Postscript	338
A Prefractionator Pinch Zone Compositions	339
B Alternative Deduction of Minimum Energy in a Petlyuk Arrangement Based on Pinch Zone Compositions	342
C Minimum Energy with a Separate Prefractionator Column	344
D Minimum Energy of a Petlyuk Arrangement based on Rigorous Simulation	348

