Article

Upcycling of Waste Poly(ethylene terephthalate) into 2,4-Pyridine Dicarboxylic Acid by a Tandem Chemo-Microbial Process

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ABSTRACT: This study presents a chemo-microbial cascade process for the upcycling of waste poly(ethylene terephthalate) (PET) into valuable compound 2,4-pyridine dicarboxylic acid (2,4-PDCA). Initially, waste PET undergoes efficient hydrolysis to terephthalic acid (TPA) with a high yield of 92.36%, catalyzed by *p*-toluenesulfonic acid (PTSA). The acid catalyst exhibits excellent reusability, maintaining activity over five cycles. Subsequently, a one-pot, two-step whole-cell conversion system utilizing genetically modified *Escherichia coli* strains (*E. coli* PCA and *E. coli* 2,4-PDCA) converts the generated TPA into 2,4-PDCA. By integrating the PET hydrolysis module with the 2,4-PDCA biosynthesis module, the study achieves an impressive overall efficiency of 94.01% in converting challenging PET waste into valuable 2,4-PDCA. Our research presents a rational design strategy for PET upcycling and 2,4-PDCA synthesis methods. This research provides a systematic approach to PET upcycling, demonstrating its feasibility and potential for industrial application.

Keywords: Poly(ethylene terephthalate); Plastic upcycling; Chemo-microbial process; 2,4-Pyridine dicarboxylic acid

Supporting Information

Figure S1. PET bottle from local supermarket

Table S1 Bacterial strains and plasmids.

Strains and plasmid	Relevant characteristic	Reference or source
Strains		
E. coli BL21 (DE3)	*E. coli str. B F– ompT gal dcm lon hsdSB(rB–mB–) λ(DE3 [lacI lacUV5-T7p07ind1 sam7 nin5]) [malB+]K-12(λS)*	
E. coli PCA(TPA to PCA)	*E. coli* BL21(DE3) with pACYCTphBTphAabc	This study
E. coli 2,4-PDCA (PCA to 2,4-PDCA)	*E. coli* BL21(DE3) with pET22b-liAB	This study
Plasmids		
pACYCTphBTphAabc	pACYC**;** T7 promoter; *TphB;TphAabc*; Cmr^R	This study
pET22b-liAB	pET22b; T7 promoter; *LigAB*; Amp^R	This study

Table S2 Primers used in this study.

Primer name	Sequence
Pacyc-TphAabc-F	CTTTAATAAGGAGATATACCATGAACCACCAGATCCATATCC
Pacyc-TphAabc-R	CCGAGCTCGAATTCGGATCCTCATAGCGGCAATGCCATC
Pacyc-TphB-F	TATAAGAAGGAGATATACATATGACAATAGTGCACCGTAGATTGG
Pacyc-TphB-R	AGCGGTGGCAGCAGCCTAGGTCAGACCGGTTGGGC
pET22b-liAB-F	CGGAATTAATTCGGATCCGATGACCGAGAAGAAAGAGAG
pET22b-liAB-R	GCAAGCTTGTCGACGGAGCTCGTCAGGCCTGGGCCAGGCTG

Table S3 Gene information used in this study.

Gene	UniProtKB Accession Number	Sequence
tphAa	Q3C1D2	ATGAACCACCAGATCCATATCCACGACTCCGATATCGC GTTCCCCTGCGCGCCCGGGCAATCCGTACTGGATGCAG CTCTGCAGGCCGGCATCGAGCTGCCCTATTCCTGCCGC AAAGGTAGCTGTGGCAACTGTGCGAGTACGCTGCTCG ACGGAAATATTGCCTCCTTCAATGGCATGGCCGTGCGA AACGAACTCTGCGCCTCGGAACAAGTGCTGCTGTGCG GCTGCACTGCAGCCAGCGATATACGTATCCACCCGAGC TCCTTTCGCCGTCTCGACCCGGAAGCCCGAAAACGTTT TACGGCCAAGGTGTACAGCAATACACTGGCGGCACCC GATGTCTCGCTGCTGCGCCTGCGCCTGCCTGTGGGCAA GCGCGCCAAATTTGAAGCCGGCCAATACCTGCTGATTC ACCTCGACGACGGGGAAAGCCGCAGCTACTCTATGGC CAATCCACCCCATGAGAGCGATGGCATCACATTGCATG TCAGGCATGTACCTGGTGGTCGCTTCAGCACTATCGTT CAGCAGTTGAAGTCTGGTGACACATTGGATATCGAACT GCCATTCGGCAGCATCGCACTGAAGCCTGATGACGCA AGGCCCCTGATTTGCGTTGCGGGTGGCACGGGATTTGC GCCCATTAAATCCGTTCTTGATGACTTAGCCAAACGCA AGGTTCAGCGCGACATCACGCTGATCTGGGGGGCTCG CAACCCCTCGGGCCTGTATCTTCCTAGCGCCATCGACA AGTGGCGCAAAGTCTGGCCACAGTTTCGCTACATTGC AGCCATCACCGACCTAGGCGATATGCCTGCGGATGCTC ACGCAGGTCGGGTGGATGACGCGCTACGCACTCACTT TGGCAACCTGCACGATCATGTGGTGCACTGCTGTGGCT CACCAGCTCTGGTTCAATCAGTGCGCACAGCCGCTTCC GATATGGGCCTGCTTGCACAGGACTTCCACGCGGATGT TTTTGCGACAGGCCCGACTGGTCACCACTAG
tphAb	Q3C1D5	ATGCAAGAATCCATCATCCAGTGGCATGGGGCCACT AATACGCGCGTGCCTTTTGGTATCTATACCGACACAG CCAATGCTGATCAGGAACAGCAGCGCATCTATCGCG GCGAGGTCTGGAACTACTTGTGCCTGGAATCTGAAA TTCCCGGGGCCGGTGATTTCCGCACTACCTTTGCCG

tphAc	Q3C1D4	GTGAAACACCGATAGTTGTCGTACGGGATGCCGACC AGGAAATCTACGCCTTCGAGAACCGCTGCGCGCATC GCGGCGCTCTCATCGCTCTGGAGAAATCGGGCCGTA CGGATAGTTTCCAGTGCGTCTATCACGCCTGGAGCT ACAACCGACAGGGAGATCTGACCGGCGTTGCCTTC GAGAAAGGTGTCAAGGGCCAGGGTGGCATGCCGGC CTCATTCTGCAAAGAAGAGCATGGCCCGCGCAAGC TCCGCGTGGCTGTCTTTTGCGGTTTGGTCTTTGGCA GTTTTTCCGAGGACGTGCCCAGCATTGAGGATTACC TTGGCCCTGAGATTTGCGAGCGCATAGAGCGCGTGC TGCACAAGCCCGTAGAAGTCATCGGTCGCTTCACGC AAAAGCTGCCTAACAACTGGAAGCTCTACTTCGAG AACGTGAAGGACAGCTATCACGCCAGCCTCCTGCAT ATGTTCTTCACCACCTTCGAGCTGAATCGCCTCTCA CAAAAAGGCGGTGTCATCGTCGACGAGTCGGGTGG CCACCATGTGAGCTATTCCATGATCGATCGCGGCGC CAAAGACGACTCGTACAAGGACCAGGCCATCCGCT CCGACAACGAGCGTTACCGGCTCAAAGATCCTAGC CTTCTAGAGGGCTTTGAGGAGTTCGAGGACGGCGT GACCCTGCAGATCCTTTCTGTGTTCCCTGGCTTTGTG CTGCAGCAGATTCAGAACAGCATCGCCGTGCGTCA GTTGCTGCCCAAGAGCATCTCCAGCTCGGAACTCAA CTGGACCTATCTTGGCTATGCAGATGACAGTGCCGA GCAACGCAAGGTCAGACTCAAACAGGCCAACCTTA TCGGCCCGGCCGGATTCATTTCCATGGAAGACGGAG CTGTCGGTGGATTCGTGCAGCGTGGCATCGCAGGCG CTGCCAACCTTGATGCAGTCATCGAGATGGGCGGAG ACCACGAAGGCTCTAGCGAGGGCCGCGCCACGGAA ACCTCGGTACGCGGCTTTTGGAAGGCCTACCGCAAG CATATGGGACAGGAGATGCAAGCATGA
tphB	Q3C1D3	ATGATCAATGAAATTCAAATCGCGGCCTTCAATGCC GCCTACGCGAAGACCATAGACAGTGATGCAATGGA GCAATGGCCAACCTTTTTCACCAAGGATTGCCACTA TTGCGTCACCAATGTCGACAACCATGATGAGGGACT TGCTGCCGGCATTGTCTGGGCGGATTCGCAGGACAT GCTCACCGACCGAATTTCTGCGCTGCGCGAAGCCA ATATCTACGAGCGCCACCGCTATCGCCATATCCTGGG TCTGCCTTCGATCCAGTCAGGCGATGCAACACAGGC CAGCGCTTCCACTCCGTTCATGGTGCTGCGCATCAT GCATACAGGGGAAACAGAGGTCTTTGCCAGCGGTG AGTACCTCGACAAATTCACCACGATCGATGGCAAGT TACGTCTGCAAGAACGCATCGCGGTTTGCGACAGC ACGGTGACGGACACGCTGATGGCATTGCCGCTATGA ATGACAATAGTGCACCGTAGATTGGCTTTGGCCATC GGCGATCCCCACGGTATTGGCCCAGAAATCGCACTG AAAGCTCTCCAGCAGCTGTCTGTCACCGAAAGGTC TCTTATCAAGGTCTATGGACCTTGGAGCGCTCTCGA GCAAGCCGCACGGGTTTGCGAAATGGAGCCGCTTC TTCAAGACATCGTTCACGAGGAAGCCGGCACACTT ACACAACCAGTTCAATGGGGAGAAATCACCCCGCA GGCTGGTCTATCTACGGTGCAATCCGCAACAGCGGC TATCCGAGCGTGCGAAAACGGCGAAGTCGATGCCG TCATTGCCTGCCCTCACCATGAAACGGCCATTCACC GCGCAGGCATAGCGTTCAGCGGCTACCCATCTTTGC TCGCCAATGTTCTTGGCATGAACGAAGACCAGGTAT TCCTGATGCTGGTGGGGGCTGGCCTGCGCATAGTGC ATGTCACTTTGCATGAAAGCGTGCGCAGCGCATTGG AGCGGCTCTCTCCTCAGTTGGTGGTCAACGCGGCGC AGGCTGCCGTGCAGACATGCACCTTACTCGGAGTGC CTAAACCAAAAGTCGCTGTATTCGGGATCAACCCTC

		ATGCATCTGAAGGACAGTTGTTCGGCCTGGAGGACT CCCAGATCACCGTTCCCGCTGTCGAGACACTGCGCA AGCGCGGCCTAGCAGTAGACGGCCCCATGGGAGCT GACATGGTTCTGGCACAGCGCAAGCACGACCTGTAT GTAGCCATGCTGCACGATCAGGGCCATATCCCCATC AAGCTGCTGGCACCTAACGGAGCCAGCGCACTATCT ATCGGTGGCAGGGTGGTGCTTTCCAGCGTGGGCCAT GGCAGCGCCATGGACATTGCCGGCCGTGGCGTGGC TGACGCCACGGCCCTCCTACGCACAATAGCCCTACT CGGAGCCCAACCGGTCTGA
lig A	P22635	ATGACCGAGAAGAAAGAGAGAATCGACGTTCACGC CTATCTCGCCGAGTTTGACGACATTCCCGGCACCCG CGTGTTCACCGCCCAGCGCGCGCGCAAGGGCTATA ATCTCAACCAGTTCGCGATGAGCCTGATGAAGGCCG AGAACCGCGAGCGGTTCAAGGCCGACGAGAGCGCC TATCTGGACGAGTGGAACCTCACGCCCGCCGCCAA GGCCGCCGTGCTGGCCCGCGACTACAATGCGATGAT CGACGAGGGCGGGAATGTCTATTTCCTGTCCAAGCT GTTCTCGACCGACGGCAAGAGCTTCCAGTTCGCCG CCGGCTCGATGACCGGCATGACTCAGGAAGAATATG CACAGATGATGATCGATGGCGGCCGTTCGCCCGCGG GTGTGCGCTCGATCAAGGGAGGCTACTGA
lig B	P22636	ATGGCACGCGTTACCACGGGCATCACGTCCAGCCAC ATTCCCGCGCTCGGCGCGGCCATCCAGACCGGCACC AGCGACAATGATTACTGGGGCCCGGTGTTCAAGGG CTACCAGCCGATCCGCGACTGGATCAAGCAGCCCG GCAACATGCCGGACGTCGTGATCCTGGTCTATAACG ACCATGCCTCGGCCTTCGACATGAACATCATCCCGA CCTTCGCGATCGGCTGCGCGGAAACGTTCAAGCCC GCCGACGAGGGATGGGGCCCGCGCCCGGTGCCCGA CGTGAAGGGCCATCCGGACCTTGCCTGGCACATCGC CCAGAGCCTGATCCTCGACGAGTTCGACATGACCAT CATGAACCAGATGGACGTCGATCATGGCTGCACCGT GCCGCTCTCGATGATCTTCGGCGAGCCCGAGGAATG GCCGTGCAAGGTCATCCCCTTCCCGGTCAATGTCGT CACTTATCCGCCGCCGTCGGGCAAGCGCTGCTTCGC GCTCGGTGACAGCATCCGCGCCGCGGTCGAGAGCT TCCCGGAAGACCTCAACGTCCATGTCTGGGGCACC GGCGGCATGAGCCACCAGCTTCAGGGCCCGCGCGC CGGCCTCATCAACAAGGAGTTCGACCTGAACTTCAT CGACAAGCTGATCAGCGACCCCGAGGAGCTGAGCA AGATGCCGCACATCCAGTATCTGCGCGAAAGCGGAT CGGAAGGCATCGAGCTGGTCATGTGGCTCATCATGC GCGGCGCGCTGCCGGAGAAGGTGCGGGATCTCTAC ACCTTCTATCACATCCCGGCCTCCAACACCGCGCTC GGCGCGATGATCCTGCAGCCGGAGGAGACCGCAGG TACACCGCTCGAACCGCGCAAGGTGATGAGCGGAC ACAGCCTGGCCCAGGCCTGA