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IS1. Highly efficient production of functional substances from synthetic compounds and secondary metabolites by mammal and microbial biotransformation

Yoshinori Asakawa[1]*

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We report herein a highly efficient production of several functional substances from synthetic aromatic compounds and plant secondary metabolites such as chalcones, mono- and sesquiterpenoids by microorganisms and rabbit. The microbial biotransformation was accomplished by four black fungi, *Aspergillus niger*, *A. sojae*, *A. usami* and *A. cellulosae* in rotatory growth cultures (120 rpm, Czapek-peptone medium at 30 °C for 3-10 days). In the case of biotransformation by mammals, each terpenoid and aromatic compound was orally administered to rabbits, and their collected urines were enzymatically treated to give each metabolite. *Aspergillus niger* cultured for 10 days in the presence of chalcone (**1**) (10 g / 200 mL medium) gave dihydrochalcone (**2**) in 95% isolated yield. The other *Aspergillus* strains converted chalcone to dihydrochalcone quantitatively. The substrate amount could be scaled up to 25 g / 200 mL medium. 4-Hydroxy- (**3**) and 4'-hydroxychalcones (**4**) incubated with the same *Aspergillus* afforded 4-hydroxydihydro- (**5**) and 3,4-dihydroxydihydrochalcones (**6**), and 4'-hydroxydihydro- (**7**) and 3',4'-dihydroxydihydrochalcone (**8**), in good yield, respectively. Thus, *Aspergillus* strains introduced a hydroxy group directly onto the already substituted benzene ring and very easily reduced the α,β-unsaturated double bond. 1,1-Diphenylmethane (**9**), 1,3-diphenylacetone (**10**), 1,3-diphenylpropane (**11**), bibenzyl (**12**), (*E* and *Z*)-stilbenes (**13, 14**) and phenylcyclohexane (**15**), grifolin (**16**), 6-shogaol (**17**), 6-gingerol (**18**), capsaicin (**19**), and dihydrocapsaicin (**20**) were also treated by *Aspergillus* strains to give the direct benzene ring hydroxylated products and ω-hydroxylation products, as well as epoxides, oxo- and hydroxyketo derivatives. (-)-α-Pinene (**21**) and β-caryophyllene (**22**) were converted by rabbits to *trans*-verbenol (**23**), an insect larvae pheromone, and 14-hydroxy-β-caryophyllene oxide (**24**), a constituent of mushrooms, in very high yield. Nootkatone (**25**), contributor to the grapefruit aroma, was obtained almost quantitatively from valencene (**26**), by a *Mucor* strain biotransformation. The metabolites of sesquiterpenoids, (+)- and (-)-cyclocolorenones (**27, 28**), (+)- and (-)-cuparenes (**29, 30**) and bisbibenzyls from liverworts, as well as the direct biotransformation of the corresponding essential oils are also reported. The presented methods are a cheap and hazard reduced eco-friendly alternative to classical organic synthesis while still reaching the high yields of the metabolites.

[1]Institute of Pharmacognosy, Tokushima Bunri University, Tokushima 770-8514, Japan.
**Corresponding author*: asakawa@ph.bunri-u.ac.jp