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## Supplementary Material

Figure 1. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM 4,5thiatryptophan. A. Selected spectra from the set of 1000 spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 345 nm (dotted line), 421 nm (dashed line), and 505 nm (solid line).

Figure 2. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM 4,5thiatryptophan in the presence of 5 mM benzimidazole. A. Selected spectra from the set of 1000 spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 345 nm (dotted line), 421 nm (dashed line), and 505 nm (solid line).

Figure 3. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM 6,7thiatrytophan. A. Selected spectra from the set of 1000 spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 346 nm (dotted line), 421 nm (dashed line), and 505 nm (solid line).

Figure 4. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM 7azatryptophan. A. Selected spectra from the set of 1000 spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 346 nm (dotted line), 421 nm (dashed line), and 498 nm (solid line).

Figure 5. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM 7azatryptophan in the presence of 5 mM benzimidazole. A. Selected spectra from the set of 1000

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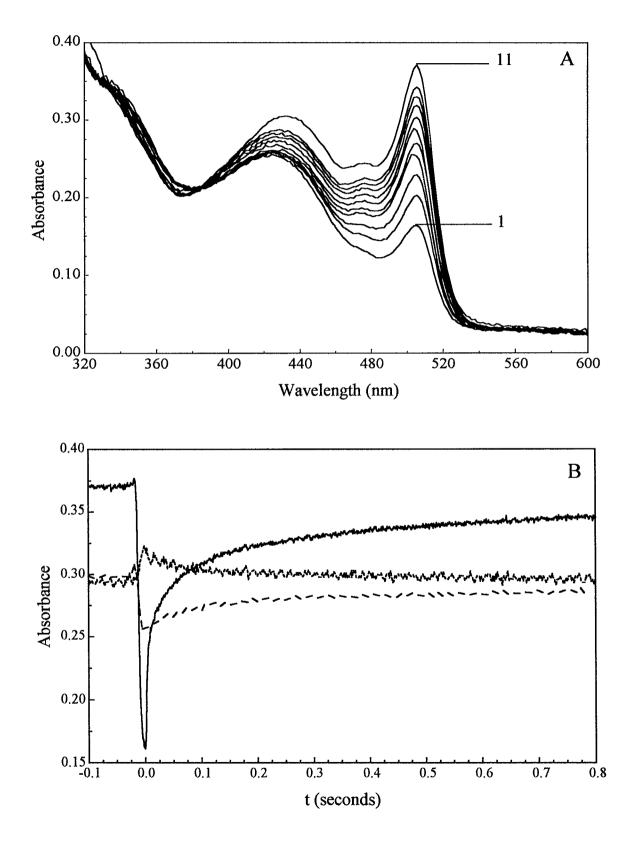
spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 346 nm (dotted line), 421 nm (dashed line), and 498 nm (solid line).

Figure 6. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM 6azatryptophan. A. Selected spectra from the set of 1000 spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 421 nm (dashed line), and 501 nm (solid line).

Figure 7. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM  $\beta$ indazolyl-L-alanine. A. Selected spectra from the set of 1000 spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 346 nm (dotted line), 421 nm (dashed line), and 501 nm (solid line).

Figure 8. Reaction of *E. coli* tryptophan indole-lyase (1 mg/mL; 19.2  $\mu$ M) with 10 mM  $\beta$ indazolyl-L-alanine in the presence of 5 mM benzimidazole. A. Selected spectra from the set of 1000 spectra collected during the reaction. Spectra shown are: 1, 0.001; 2, 0.003; 3, 0.005; 4, 0.010; 5, 0.020; 6, 0.040; 7, 0.080; 8, 0.160; 9, 0.320; 10, 0.640 seconds, and 11, steady-state (approximately 1 minute after the start of the reaction). B. Time courses for the reaction taken at 346 nm (dotted line), 421 nm (dashed line), and 501 nm (solid line).





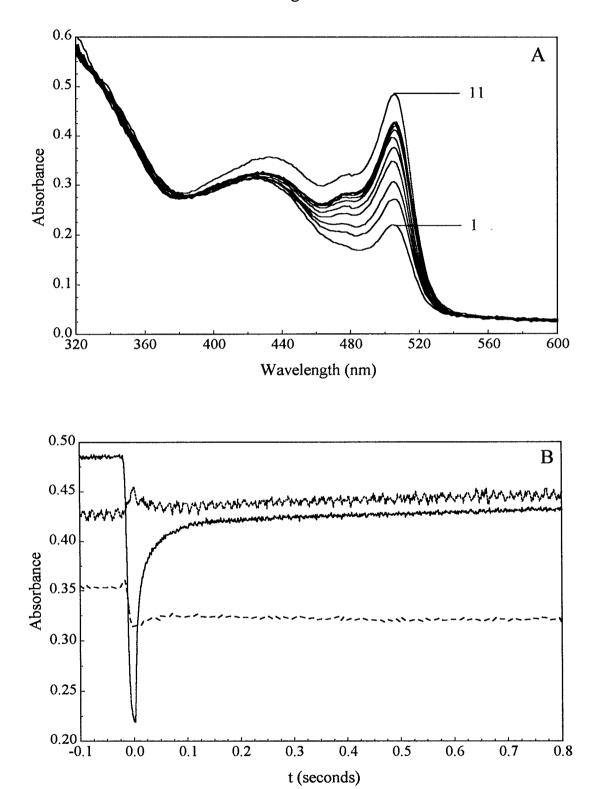
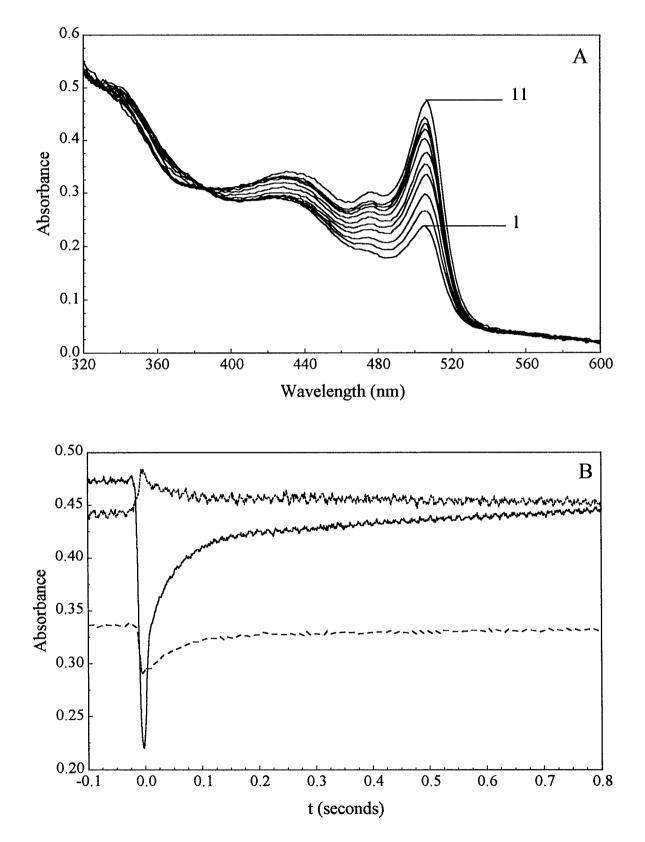


Figure 2





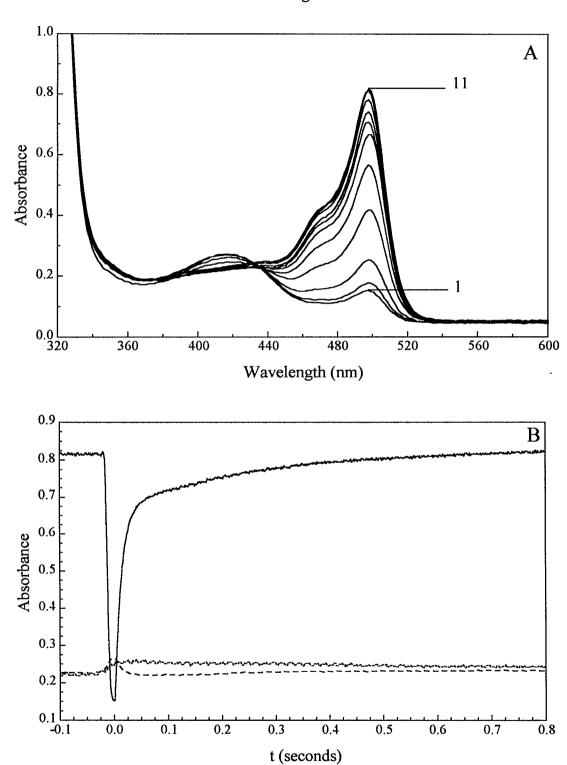
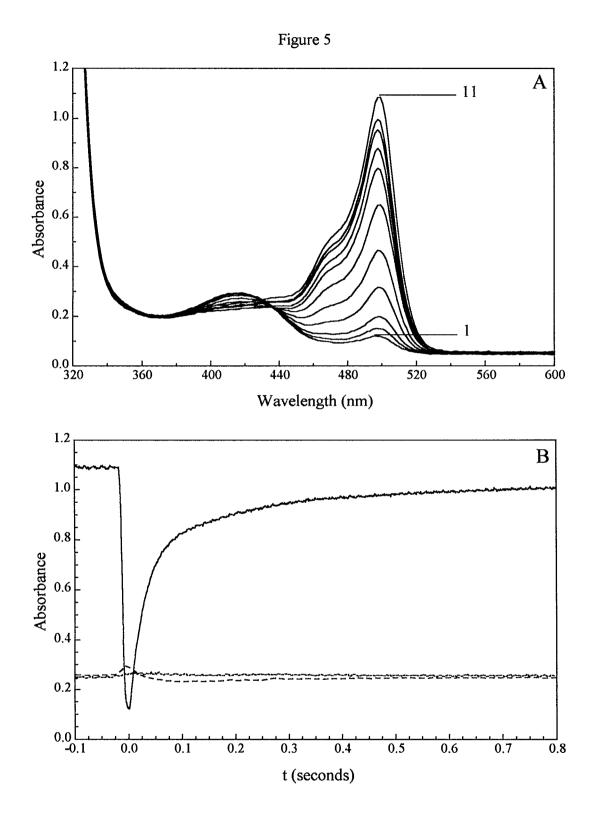
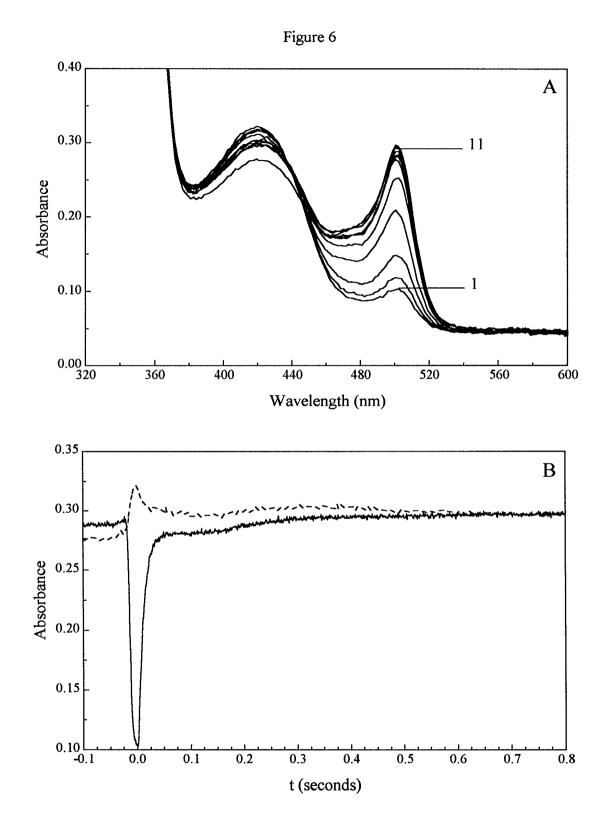


Figure 4





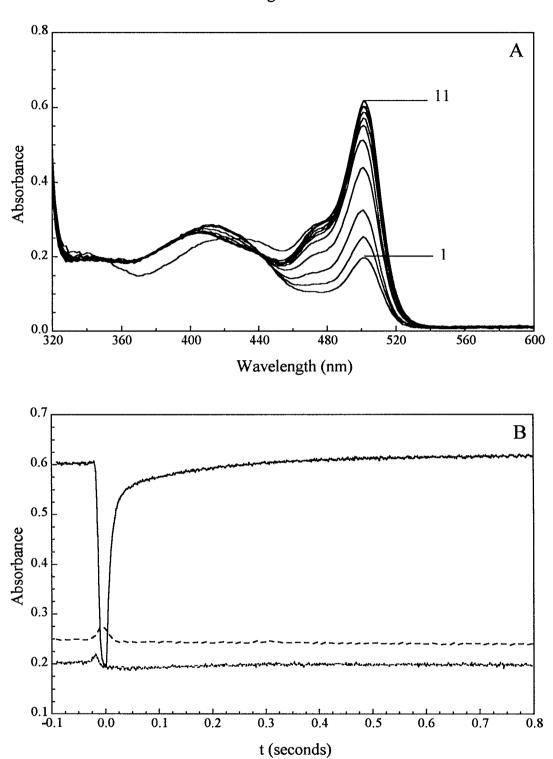


Figure 7

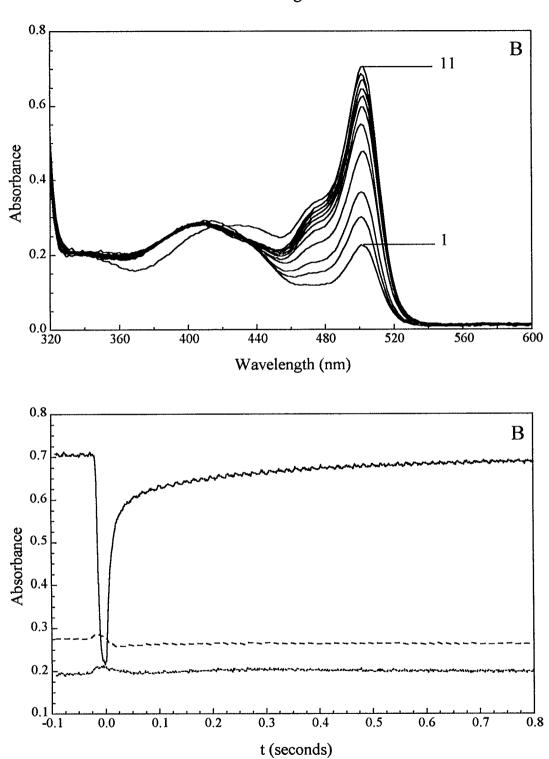


Figure 8