Supporting Information

for

Degradation of Lignin by Infrared Free Electron Laser

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1. IR-FEL spectra at several wavelengths.

IR-FEL spectra tuned to 6.3 μm, 7.1 μm, and 3.0 μm were shown in **Fig. S1**. Each spectrum was obtained by introducing the beam line into the spectrometer. Two mid-infrared FEL spectra at 6.3 μm and 7.1 μm were measured at KU-FEL (Kyoto University), and one near-infrared FEL spectrum at 3.0 μm was measured at LEBRA (Nihon University).

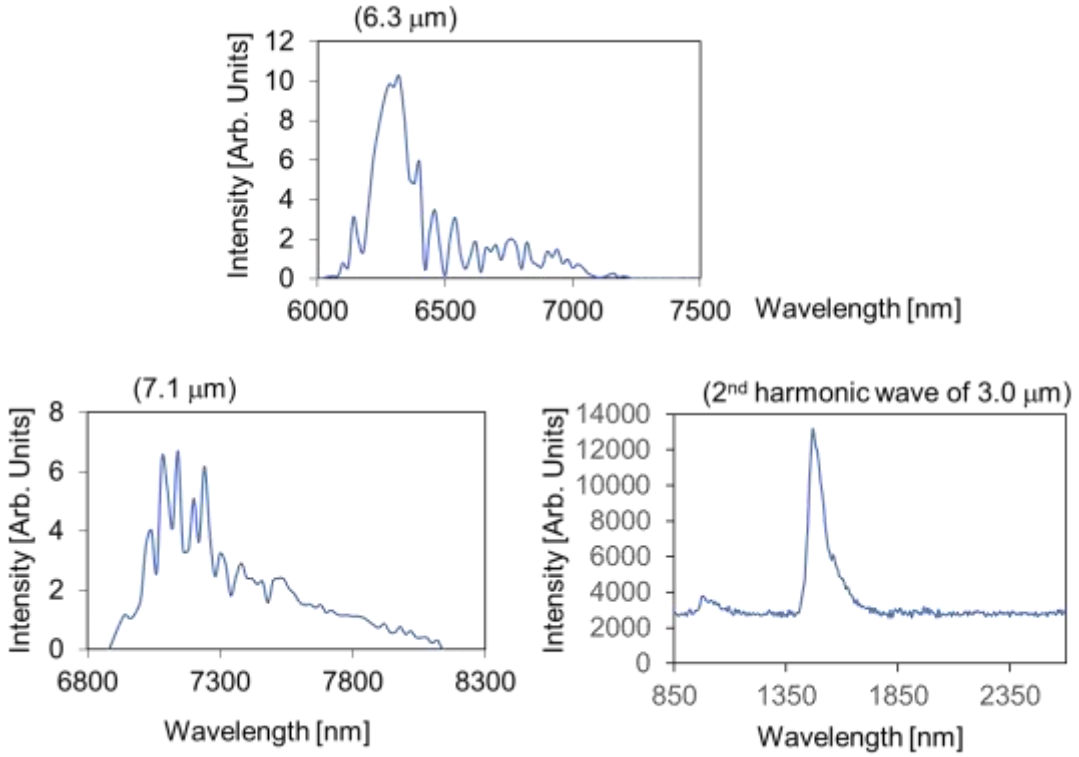
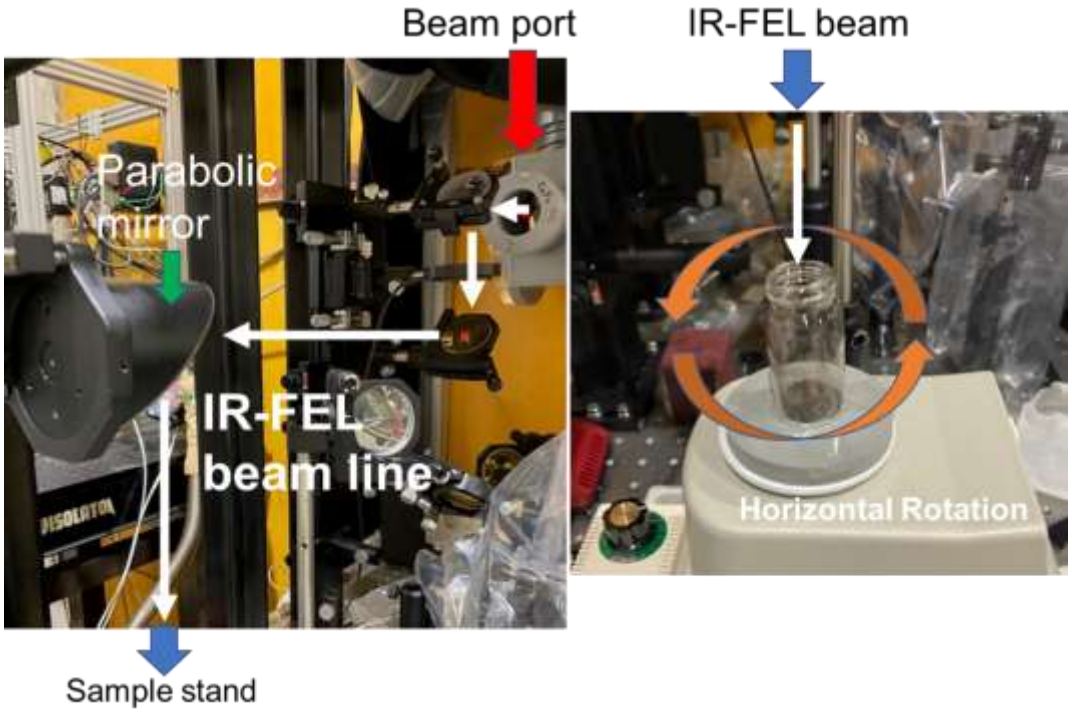


Figure S1. FEL spectrum at 3.0-7.1 μm. The near-IR FEL spectrum was measured as a 2nd harmonic wave (1475 nm).

2. Laser irradiation method

The laser beam was introduced from a beam port to the sample surface by using two reflection mirrors and one parabolic mirror in the case of mid-IR FEL system (**Fig. S2**, upper). In the case of near-IR FEL system (below), the beam direction was controlled by using three reflection mirrors and introduced onto the sample by using a focused lens (CaF_2, f = 100 mm, diameter: 50 2

mm). The lignin powder (about 5 mg) was added in a glass tube, and the IR-FEL was irradiated from the vertical direction at room temperature under atmospheric conditions. The glass tube was sometimes shaken horizontally to give the radiation energy to the whole sample uniformly.



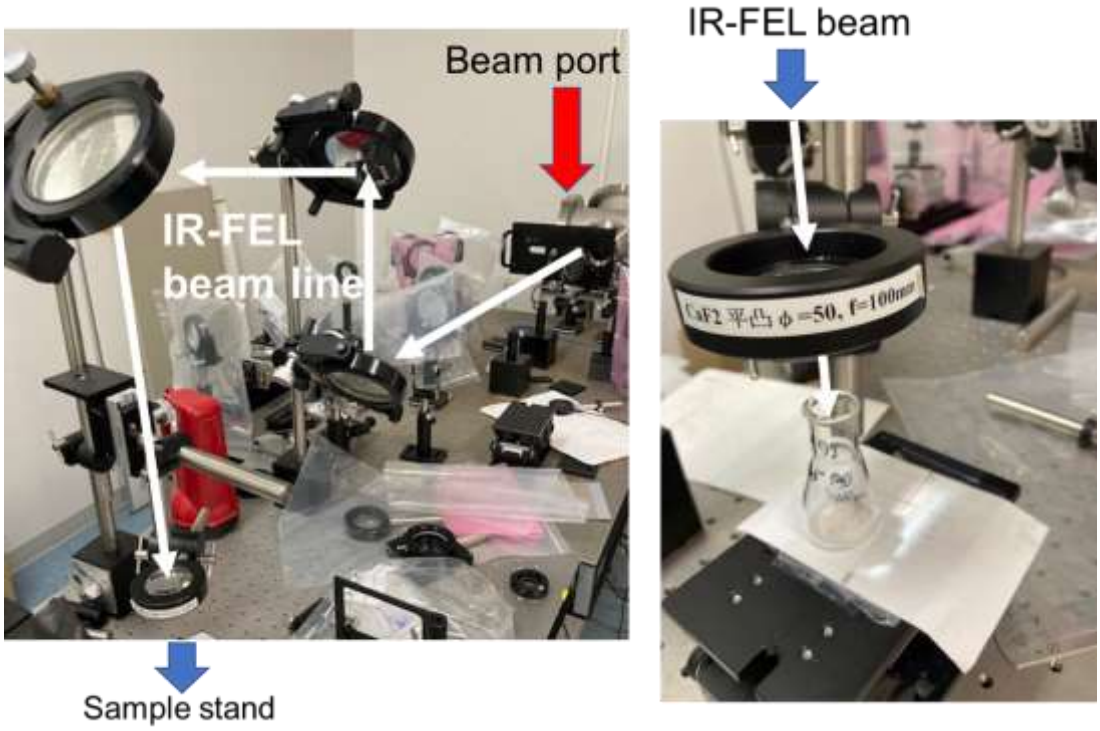


Figure S2. Experimental setup. Upper: mid-IR FEL irradiation system; below: near-IR FEL irradiation system.