It is known since decades that radiolytic splitting of water results in the formation of various radicals, which eventually form H₂O₂ and H₂. Interestingly, with the exception of radical-initiated polymerization of vinyl monomers or hydrogen production, this reaction has not yet caught the attention of organic chemists. Particularly, hydrogen peroxide could be used to drive a broad range of catalytic oxidation reactions. Peroxygenases (UPOs, E.C. 1.11.2.1), for example, are a class of enzymes catalyzing a broad range of specific, H₂O₂-dependent oxyfunctionalization reactions ranging from the hydroxylation of aromatic and aliphatic C–H-bonds, epoxidation of C=C-bonds, and oxygenation of heteroatoms. For this, peroxygenases utilize a heme prosthetic group, which in the presence of H₂O₂ is transformed into an oxo-ferryl species (Compound I) mediating the oxyfunctionalization reaction (Scheme 1). Utilizing this "H₂O₂ shunt pathway", peroxygenases are independent from the complex electron transport chains utilized by P450 monooxygenases to form Cpd I via reductive activation of O₂.

In the presence of too high concentrations of H₂O₂, however, peroxygenases are also irreversibly inactivated. To alleviate this issue, a range of in situ H₂O₂ generation methods have been developed, mostly comprising catalytic reduction of H₂O₂. These systems can be categorized by the sacrificial reductant used (Table S1). The well-known glucose oxidase system, for example, transforms glucose into gluconic acid, thereby yielding more than 190 g of waste per mol H₂O₂ generated. Formic acid, methanol, H₃PO₄, or electrochemical power are more attractive from the atom economy point of view. Water oxidation appears most appealing as here, the atom efficiency is the highest. In this context, the radiolytic formation of H₂O₂ may represent an interesting alternative method (Figure 1a).
As a radiation source, we used an external gamma radiation source ⁶⁰Co, which is widely applied, for example, in radiotherapy (i.e., gamma knife) and sterilization.

Indeed, an aqueous buffer placed next to the radiation source steadily accumulated H₂O₂ up to 0.1 mM at which the H₂O₂ concentration plateaued (in the case of a dose rate of 12.9 Gy min⁻¹) (Figure 1b). In another experiment, we presupplemented the buffer with 0.5 mM H₂O₂ and observed a steady decrease in the H₂O₂ concentration to approximately 0.1 mM (Figure 1b). Apparently, the constant H₂O₂ concentration was the result of a steady state between H₂O splitting (yielding H₂O and O₂) and radiolysis-based splitting of H₂O₂ (yielding H₂O and O₂). The position of the steady state depended on the intensity (i.e., dose rate) of the radiation source (Figure S1).

Next, we combined the ⁶⁰Co-induced water radiolysis with a UPO-catalyzed hydroxylation reaction. As a model reaction, we used the selective hydroxylation of ethyl benzene to (R)-1-phenyl ethanol catalyzed by the recombinant, evolved peroxygenase from Agrocybe aegerita (rAaeUPO). To confirm that the overall reaction followed the mechanism outlined in Figure 2a, a range of control reactions were executed: performing the reaction either in the absence or using thermally inactivated rAaeUPO yielded no product formation, while in the presence of rAaeUPO, enantiomerically pure (>99% ee) (R)-1-phenyl ethanol was formed. The presence or absence of molecular oxygen had no obvious influence on the product formation rate. Furthermore, performing the reaction in H₂¹⁸O-labeled buffer resulted in the formation of (R)-1-phenyl ethanol (Figure 2b,c). This confirms that indeed the reaction medium serves as a source of H₂O₂ and that reduction of O₂ (from ambient air) played a minor role in the H₂O₂ formation.

(R)-1-phenyl ethanol was the sole product observed, indicating that the selectivity of the biocatalyst was not impaired under the reaction conditions, particularly by the radioactivity. A control reaction with (R)-1-phenyl ethanol only under the irradiation showed that radiation-induced further oxidation of the primary enzyme product ((R)-1-phenyl ethanol) can be ruled out.

Next, we further investigated some factors influencing the efficiency and robustness of the overall reaction (Figure 3). Increasing the biocatalyst concentration increased the product formation within the first hour (Figure 3a). This increase, however, was not linear and converged to approx. 0.25 mM h⁻¹ at rAaeUPO concentrations above 100 nM. Interestingly, this product formation rate was approx. twofold higher than the H₂O₂ accumulation rate observed in the absence of the biocatalysts (Figure 1b). This observation can be attributed to the irreversible peroxygenase step removing H₂O₂ from the steady-state equilibrium. A respectable turnover number for the biocatalyst (TN = moles(rAaeUPO) × moles[(R)-1-phenyl ethanol]) of more than 1400 was observed for the biocatalyst.

These experiments, however, also revealed a poor long-term stability of the enzyme under the reaction conditions. Already after 1 h of reaction (approx. 770 Gy under the dose rate of 12.9 Gy min⁻¹), the product formation ceased, which we interpreted as loss of enzyme activity (Figure S2). This assumption is supported by a considerable decrease in the
As major contributors to the observed biocatalyst inactivation, we tested a range of different radical scavengers (Figure 3b). Among these radical scavengers especially methanol, acrylamide, and formate enabled significantly increased product formation (Figure 3b). The effect depended on the concentration of the radical scavenger as exemplified with methanol and formate (Figures S5 and S6). We therefore also compared the time courses of the radioenzymatic reactions in the absence and presence of the radical scavengers methanol and formate (Figure 3c). Most strikingly, the conversion of ethyl benzene to (R)-1-phenyl ethanol was increased from approx. 18%, in the case of reactions in the absence of radical scavengers, to full conversion, in the presence of sodium formate. In the latter case, a turnover number for the biocatalyst of 40.000 was achieved, which we ascribe to a higher enzyme stability because of a decreased concentration of hydroxyl radicals. This assumption was also supported by electron paramagnetic resonance experiments, which revealed that in the presence of both methanol or formate, the in situ \( \cdot \)OH concentration was significantly reduced (Figures S7 and S8).

The dose rate of the radiation source directly influenced the product formation of the radioenzymatic reaction system (Table 1). The final product concentration (and directly related to this also the turnover number of the enzyme) directly correlated with the dose rate of the radioactivity source applied. Interestingly, the “radiation yield”, that is, the amount of product formed per Gy, correlated inversely with the dose rate. This may be due to a decreased radiolytic \( \cdot \)H\(_2\)O\(_2\) decomposition at lower dose rates, whereas the biocatalyst concentration remained constant. Further experiments will be necessary to fully rationalize this observation. Pleasantly, the reactions performed with spent fuel element \( ^{235}\)U also showed good robustness.

Table 1. Radioenzymatic Hydroxylation of Ethyl Benzene Using Different Radiation Sources

| radiation source \(^b\) | \(^{60}\)Co-1 | \(^{60}\)Co-2 | \(^{235}\)U |
|--------------------------|----------------|----------------|----------------|
| dose rate \([\text{Gy m}^{-1}]\) | 12.9 | 1.0 | 1.67 |
| (R)-1-phenyl ethanol \([\text{mM}]\) | 0.91 | 0.29 | 0.39 |
| ee [\%] | >99 | >99 | >99 |
| TON\(_{\text{AaeUPO}}\) \(^d\) | 18,200 | 5800 | 7800 |
| radiation yield \([\mu \text{M}_\text{product} \times \text{Gy}^{-1}]\) | 0.196 | 0.806 | 0.659 |

\(^{\text{a}}\)General reaction conditions: sodium phosphate buffer (60 mM, pH 7), [ethyl benzene] = 1 mM, [\(\text{AaeUPO}\)] = 50 nM, [sodium formate] = 50 mM, \(T = 22 \, ^{\circ}\)C, \(t = 1 \text{ h}\). \(^{\text{b}}\)Three different radiation sources were used: \(^{60}\)Co-1 and -2 exhibiting dose rates of 12.9 and 1 Gy min\(^{-1}\), respectively, and \(^{235}\)U (from a spent fuel element) with 1.67 Gy min\(^{-1}\). \(^{\text{c}}\)TON and \(^{\text{d}}\)radiation yield = product concentration \(\times\) (dose rate \(\times\) reaction time)\(^{-1}\).
Experiments were performed as duplicates; (a) [substrate] = 1 mM, cosolvent, [tridecanoic acid (50 mM Tris-HCl, pH 8), 30% (v/v) MeCN as citrate buffer], 42,43 significant product accumulation was observed. Possibly, the radicals present in the reaction mixture enabled “donor-independent” H2O2 generation from water. The dose-rate-dependent steady-state concentration appears ideal to provide heme-dependent peroxygenases with suitable concentrations of H2O2 that enable the reaction while minimizing the oxidative inactivation. This advantage, at least in the present setup, is compensated by the radical-induced inactivation of the biocatalyst, this is also reflected by the comparably poor performance of the present system compared to other in situ H2O2 generation systems (Table S1). Compared with (enzymatic) H2O2 generation systems (which largely avoid the intermediate occurrence of radical species), the peroxygenases’ turnover numbers fall back approx. 10-fold. Compared to other (radical-generating) H2O2 generation systems, the turnover numbers observed here compare very well. The radical inactivation of the biocatalysts represents an apparent shortcoming of the current setup. In future experiments, we will address this by physical separation of the biocatalyst from the radiation source. Flow chemistry appears a particularly attractive technical solution.

Although this approach at first sight may appear as a lab curiosity, we believe that it may actually bear some practical relevance. In this study, we have demonstrated that spent fuel elements can drive peroxygenase-catalyzed reactions. Considering the annually increasing amounts of radioactive waste and its persistence, the proposed radioenzymatic approach may represent a possibility to productively utilize nuclear waste. Furthermore, it should be kept in mind that globally a variety of different radiation sources are used commercially. For instance, 60Co units are used for sterilization and electron beams for various applications and research nuclear reactors (more than 250 worldwide).

### EXPERIMENTAL SECTION

#### Production of the Biocatalysts.

The evolved, unspecific peroxygenase from Agrocybe aegrita (rAaeUPO) was obtained from fermentation of recombinant Pichia pastoris as previously described.35,34 The culture broth containing rAaeUPO in the supernatant was clarified by centrifugation followed by ultrafiltration and filtered through a 20 μm filter. The enzyme preparation was stored at −80 °C until further use. The vanadium-dependent chloroperoxidase from C. inaequalis (CVCPO) was produced by recombinant expression in Escherichia coli as described previously.35 The crude cell extracts were treated with isopropanol (50% v/v) to precipitate nucleic acids and endogeneous E. coli proteins. The clarified supernatant was supplemented with (NH4)2VO4 (100 μMfinal) to reconstitute the holoenzyme.

Radiochemical Experiments. All radiochemical experiments were performed by placing 2 mL GC vials filled with 1 mL of the reaction mixture next to the radioactivity source (Figure S10). All reactions were performed at ambient temperature (22 °C). At intervals, samples were removed from the radiation source and analyzed. For H2O2 quantification, we used using Ghormley’s triiodide method.44 For the analysis of the radioenzymatic reactions, the reaction mixtures were further processed and analyzed by GC or HPLC as described previously.21,45,46

### ASSOCIATED CONTENT

+ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acscatal.0c03059.

Detailed experimental and analytical details and further experimental results (PDF)
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Notes
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