Polymer gel dosimeters have been developed for three dimensional radiation dosimetry using MRI. However, the mechanisms determining the dose dependence of the polymer gel NMR spin-relaxation are still under study. Here we report the results of a basic investigation of a polymer gel dosimeter (3% bis, 3% acrylamide, and 5% gelatin) irradiated to doses from 0 to 10 Gy. Spin-spin relaxation (R2) and rotating frame spin-lattice relaxation (R1p) rate measurements were performed at 5 °C using CPMG and 10 G spin-locking pulse sequences, respectively. Both spin-spin and rotating frame spin-lattice relaxation decays are bi-exponential with fast and slower relaxing magnetization components. At zero dose the magnetization fraction of the fast decaying component is ~4% in both R2 and R1p experiments; this corresponds stoichiometrically to the signal expected from the gelatin in the dosimeter before the onset of radiation induced polymerization. The magnetization fraction of this component increases linearly with dose with a slope of ~0.5 percent per Gy, enabling the determination of an effective chemical yield for this polymer gel dosimeter. All R2 and R1p rates increase linearly with dose, with the dose sensitivity of the relaxation rates associated with gelatin and polymer protons about three times greater than that of the water/monomer rates. This is expected since the effect of polymerization on the water proton component is indirect, mediated by protons on water hydrating the polymer molecules. The practical implications of the newly observed two component magnetization decay for MRI polymer gel dosimetry techniques will also be discussed.