Since the advent of gaseous nitriding and nitrocarburizing processes in the early 20th century, numerous process variants and applications have been developed. Improved performance with respect to fatigue, wear and corrosion can be obtained for treated steel components. Despite the advantages and widespread application, the current understanding of the thermodynamics and kinetics governing the processes, as well as the resulting crystal structures and magnetic properties, is still far from complete. An incomplete fundamental description hinders possible process optimization and has motivated the current work in synthesis and characterization of interstitial solutions of nitrogen and carbon in iron-based lattices. In order to avoid the influences of gradients in composition and residual stresses, which are typically found in treated surface layers, homogeneous samples are needed. These were prepared from pure iron or austenitic stainless steel using gaseous mixtures of ammonia, hydrogen, acetylene and propene at elevated temperatures. Structural and magnetic properties have been characterized with neutron diffraction, vibrating sample magnetometry and Mössbauer spectroscopy. Thermal decomposition and decomposition was studied in inert atmospheres with in situ synchrotron X-ray diffraction. Thermal decomposition sequences for iron carbides and carbonitrides, as well as for so-called expanded austenite, were established. In ε-iron nitride, partial substitution of nitrogen by carbon causes an increase in Curie temperature and specific magnetization. Changes in interstitial ordering were deduced from the observed effects on lattice parameters and related to relatively favorable interactions between nitrogen and carbon.

At room temperature expanded austenite is found to be paramagnetic for high and low nitrogen contents but ferromagnetic for intermediate contents. An anomalous variation in thermal expansion coefficients with interstitial content is caused by spontaneous volume magnetostriction in the ferromagnetic state.

Several structural models have previously been proposed for expanded austenite, but all have failed to successfully describe all features of X-ray diffraction data. In the current evaluation of diffraction data, the effects of stacking faults on hkl-dependent shifts of Bragg reflections were included. Comparison of simulations with experimental data does, however, show that the stacking fault model is not entirely acceptable. The apparent anisotropic deviations from cubic symmetry currently lack an exact quantitative interpretation.