Silicone rubber is one of the most important types of synthetic rubbers with good heat resistance but poor thermal conductivity. In order to further increase the good heat resistance and improve thermal conductivity of silicone rubbers, inorganic fillers such as Fe2O3 are used. However, Fe2O3 particles are easy to agglomerate because of their high surface energy. The poor compatibility between Fe2O3 and silicone rubber results in poor dispersion of Fe2O3 particles within the silicone rubber matrix. The bad dispersion of Fe2O3 particles restricts the improvement of thermal conductivity by addition of Fe2O3 particles. In this study the surface of Fe2O3 particles were modified with vinyltrimethoxysilane (VTMS) by means of wet chemical surface modification to improve dispersion state of Fe2O3 particles and consequently thermal properties of silicone rubber. VTMS is a surface modifying agent containing methoxy and unsaturated vinyl groups. The methoxy group can hydrolyze to form hydroxyl group and react with active groups on the surface of Fe2O3. On the other hand, vinyl group may take part in the crosslinking process of silicone rubber which is beneficial to a good interfacial bonding between Fe2O3 particles and silicone rubber. FTIR analysis showed that VTMS was attached onto the surface of Fe2O3. The effect of different amounts of the modified Fe2O3 on the mechanical properties before and after ageing, thermal conductivity and heat resistance of silicone rubber samples were investigated. A better dispersion of Fe2O3 particles within the matrix was observed which led to an improvement in the thermal conductivity higher than that of silicone rubber containing unmodified Fe2O3 particles. The observed improvement in the dispersion of modified Fe2O3 particles in silicone rubber and the decrease in their aggregation was attributed to the increased interactions between modified Fe2O3 particles and silicone rubber.