The gravest major sins are: associating partners with Allah in worship, feeling secure from Allah's plan, despairing of Allah's mercy, and losing hope in relief coming from Allah

‘Abdullāh ibn Mas‘ūd (may Allah be pleased with him) reported that the Prophet (may Allah's peace and blessings be upon him) said: "The gravest major sins are: associating partners with Allah in worship, feeling secure from Allah's plan, despairing of Allah's mercy, and losing hope in relief coming from Allah.”

[Its chain of narrators is Sahih/Authentic] [‘Abdur-Razzaaq]

The Prophet (may Allah's peace and blessings be upon him) mentioned in this Hadīth the sins that are considered the gravest and worst ones. These sins include associating partners with Allah, the Almighty, in His lordship or His worship. He started with polytheism because it is the gravest sin ever. Another sin is losing hope in Allah’s mercy, because it involves having ill expectations of Allah, and ignorance of the vastness of His mercy. It is one of the gravest sins as well to feel secure against being dragged by way of favors until Allah punishes the sinner all of a sudden. The Hadīth, however, does not aim at listing all the major sins, rather, it aims at highlighting the gravest of them, because the list of major sins contains so many more.

http://www.sunnah.global/hadeeth/en/show/6049