The energy and chemical sectors are suffering remarkable changes. In this way, producing abundant quantity of energy with high quality, economic and environmental viability and sustainability is the main concern of the present times. One of such vital components of the world’s energy supply that fulfills the abovementioned requirements is natural gas [1].

Natural gas consists of 85–95% methane, but it also contains considerable amounts of heavier hydrocarbons as well as other compounds (CO₂, N₂, Hg, He, H₂S) [1]. In order to meet the pipeline quality standard specifications or Liquefied Natural Gas (LNG) production, impurities must be removed. Carbon dioxide is one of the major contaminants in natural gas, reducing its energy content and becoming acidic and corrosive in the presence of water, damaging pipelines and equipment. In LNG processing plants CO₂ can solidify, blocking the pipeline systems and causing transportation drawbacks [1]. In order to meet the pipeline quality standard specifications an upper limit for nitrogen and carbon dioxide of 4 and 2% is necessary, respectively. While, to inhibit the formation of dry ice in the liquefaction step, CO₂ content must be reduced to 50 ppm level [2, 3].

Among the several separation technologies available for natural gas upgrade, such as chemical absorption, physical absorption, cryogenic distillation and membrane processes, adsorption processes are considered a competitive solution. Removal of carbon dioxide from natural gas for LNG production, can be carried out by cryogenic distillation using a multicolumn sequence to obtain a product stream with less than 50 ppm of CO₂ [4]. Cryogenic adsorption-based processes, particularly pressure thermal swing adsorption, arise as a reliable and innovative alternative to replace the distillation columns, in particular the last one.

In this work, the implementation of a new methodology to design and simulate an industrial scale pressure and temperature swing adsorption process at low temperature (Cryo-PTSA) is presented, in order to produce an almost pure methane stream, with a maximum of 50 ppm of CO₂ from a stream containing 0.9% of CO₂, maximizing the methane recovery. Binderless 13X zeolite was selected as adsorbent and adsorption equilibrium isotherms of methane and carbon dioxide were determined experimentally, in the temperature range 183–473 K, up to 80 bar. The GERG-2008 Equation of State (EoS) was used during the dynamic simulations to account for deviation from ideal gas law, through the use of REFPROP (REFerence fluid PROPerties) software package. A methane recovery of 90.7% and a product stream with 41.8 ppm of CO₂ in CH₄ is achieved after cyclic steady state is reached. Cryo-PTSA simulation results were then compared with an existing cryogenic distillation process. The results obtained by process simulation showed that it is possible to replace the 3rd column of the cryogenic distillation process by a Cryo-PTSA achieving higher CH₄ recovery and lower power consumption (2.2 MW).