Regeneration of “PU” palladium catalyst with the use of supercritical fluid extraction.

Tyapkin E., Yarullin L., Gabitov F., Gumerov F.
Russia, Kazan State Technological University

The purpose of this research was to carry out regeneration of palladium catalyst with the use of supercritical carbon dioxide as an extracting agent, modified by polar substance.

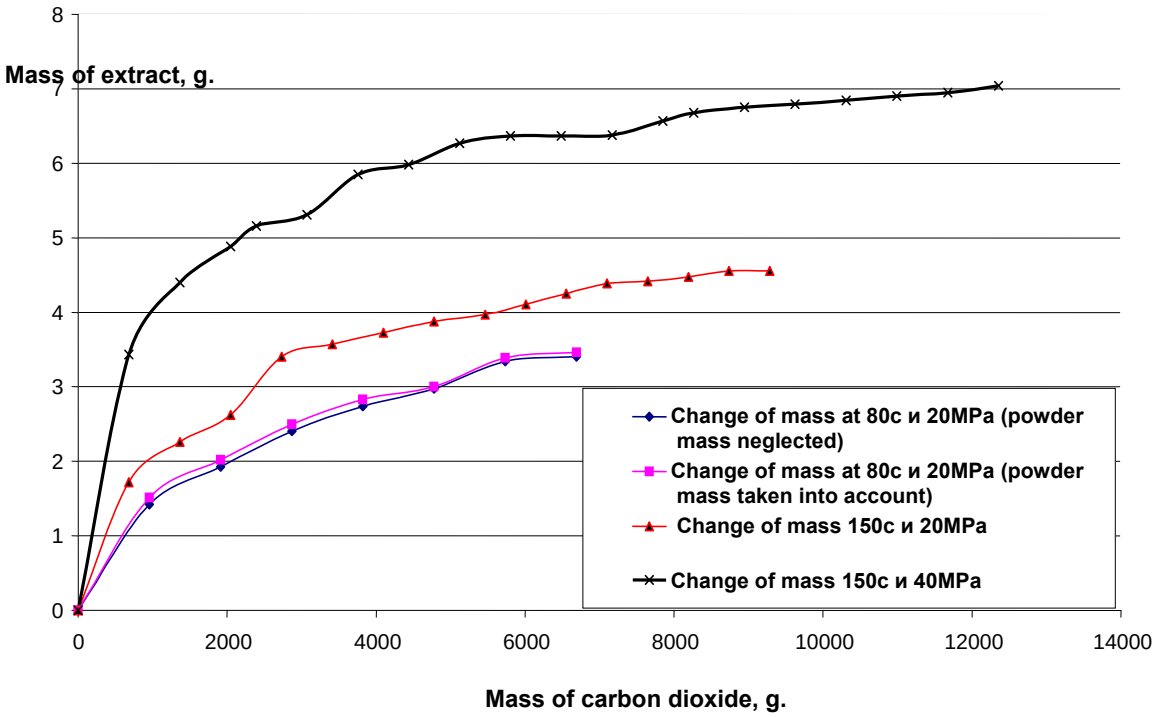


Figure1.Change of catalyst mass as a function of amount of carbon dioxide

The results of the experiments performed at different temperatures and pressures are shown in figures 1 and 2.

Maximum value of purification obtained at 20 MPa using 10 kg of CO_2, may be reached at 40 MPa at a shorter time consuming only 1.5 kg of CO_2. Moreover, at 40 MPa even better extraction results may be observed.

A waste ethylene hydrogenation catalyst pellets are shown in figure 2a (zoomed by 15). The surface of the catalyst is glossy.

In figure 2b purified catalyst pellets regenerated at 20 MPa and 150 ^{0}C are shown. Here the surface of the catalyst is black and dull, but with a thin layer of residual coke.

In figure 2c catalyst pellets regenerated at 40MPa and 150^{0}C are shown. No residual coke is observed, the surface is dull and dark-grey colored. Also metallic particles – palladium grains – may be seen.

During the separation stage sufficient amounts of deactivating substances, carbon and hydrogen containing compounds, were obtained (dark tarry fluid).

Fluid and powder, which were extracted during regeneration, are shown in figures 2d and 2e. The atomic composition data for extracted compounds, obtained by means of infrared spectrometry (tables 1 and 2) makes it possible to suppose that compounds, extracted from catalyst, were coke and polymer substances.

Atomic composition of fluid

Table 1

Element	*C*	*H*	*N*	*S*	*Cl*	*Pd*
Percentage by mass	*80,58*	*3,56*	-	*0,85*	*1,4*	-

Atomic composition of powder

Table 2

Element	*C*	*H*	*N*	*S*	*Cl*	*Pd*
Percentage by mass	*74.36*	*7.91*	-	-	*0.94*	*8.77*

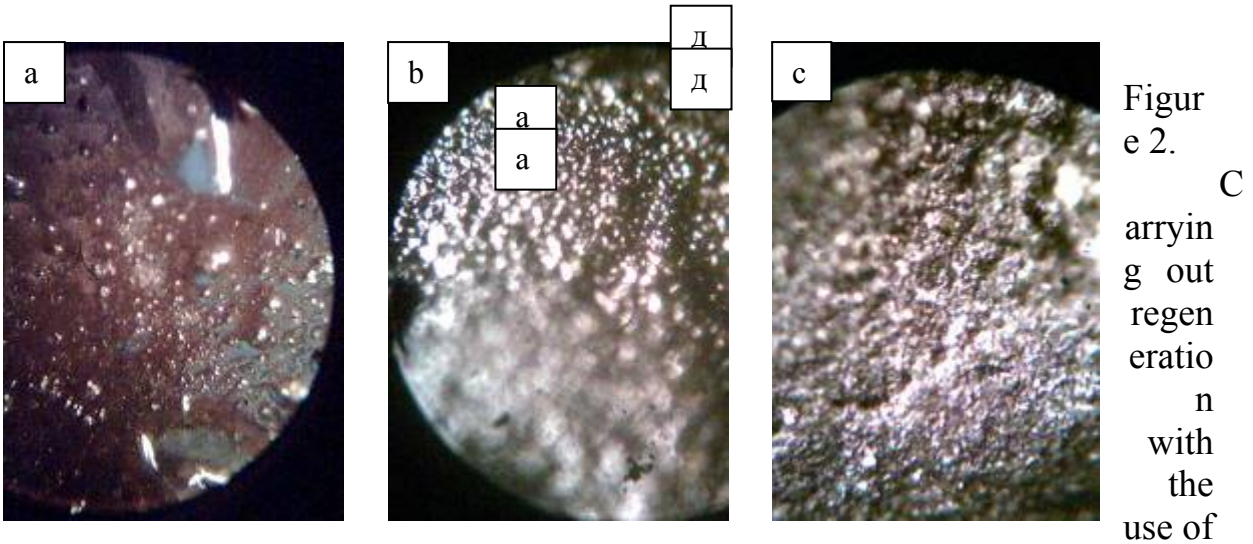
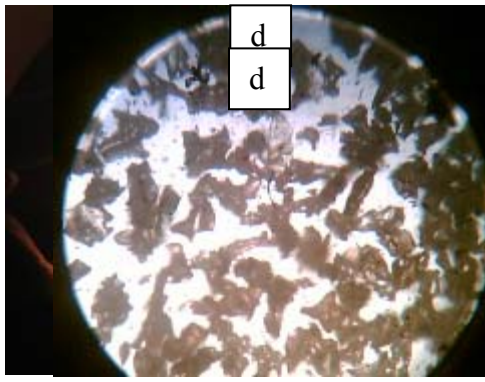


Figure 2. Carrying out regeneration with the use of carbon dioxide at high pressure (40 MPa) and temperature 150^0C enables almost complete removal of catalyst deactivating substances.



During the study of palladium catalyst regeneration process the comparative effectiveness of different supercritical extractant flow directions in the extractor were examined (figure 3).

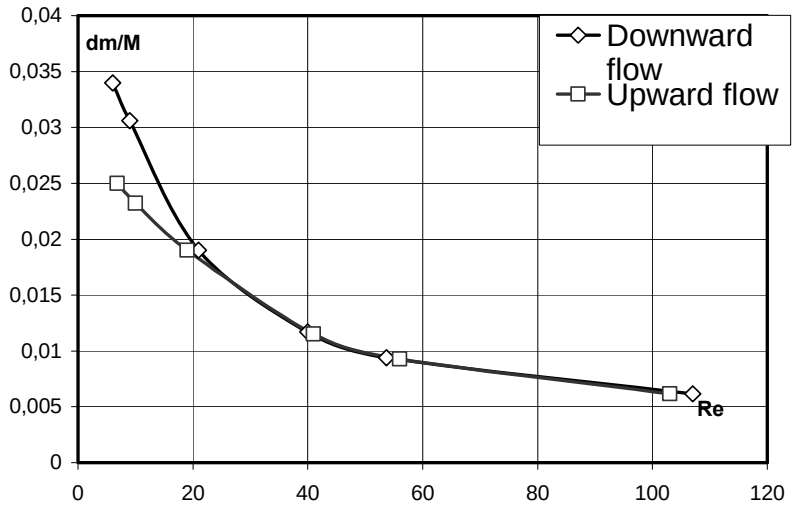


Figure 3. The change of palladium catalyst mass as a function Re criterion at different extractant flow directions are shown.

A propane-butane mixture, containing of equal masses of both hydrocarbons, has been used as an extractant. The process was carried out at $T=150^0C$ and P=6MPa. At low Re criterion values (Re<20) it is more appropriate to provide an upward flow of extractant.