

Positron Annihilation Analysis for Zeolites/Silica Gel used in Catalysis

J.M.Urban-Klaehn^{1*}, C.N. Taylor¹, K.L. Gering¹, J.D. Rimer², and R.R. Fushimi¹

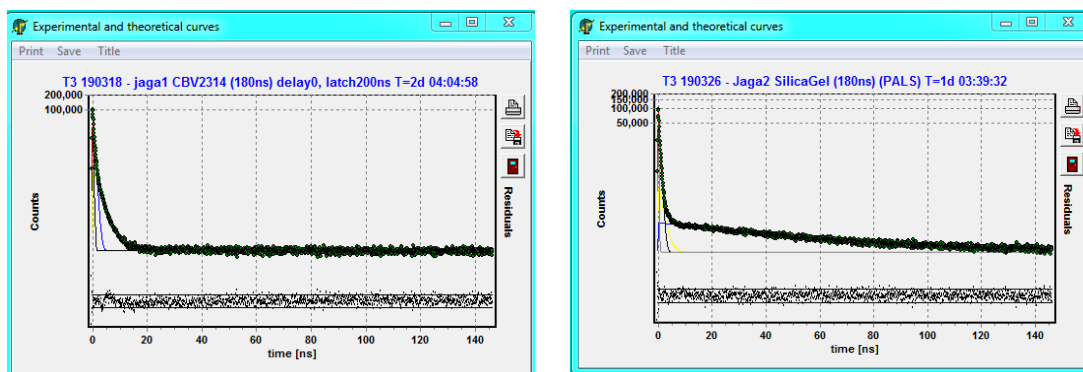
¹Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415 US

²University of Houston, Chem/Biom. Engineering Dept., 4726 Calhoun Road, Houston TX US

Pelletized samples of zeolite (H-ZSM-5), silica gel and their mixture used in catalytic hydrocarbon processing, were investigated using Positron Annihilation Lifetime (PAL) and Coincidence Doppler Broadening (CDB). In the catalytic testing of methanol conversion to olefins it is necessary to dilute the H-ZSM-5 (catalyst) with inert silica to achieve more uniform changes in the material with time-on-stream, therefore the Zeolite/silica gel ratio equal 1: 3.054 for the mixture was used.

The PAL measurements were done in two different time regimes up to 150 ns to detect long lifetimes due to a Positronium (Ps) formation. The surface of the pelletized and non-pelletized (powder) samples were examined under an optical digital microscope to identify the effects of pelletizing, and the homogeneity of the mixture.

Fig. 1 PAL for H-ZSM-5 (left) & silica gel (right) by use of LT-9.2 program [1], extended time scale



Zeolites represent a class of porous minerals with uniform, regular and highly organized micro-structure. The PAL measurements showed 2.4 ns lifetime due to micro-pores: 0.3-0.4 nm diameter [2], with the intensity over 20% for pure zeolite, about half of this intensity for the mix, and none for the pure silica gel [Fig.1]. On the contrary, silica gel exhibited a 44 ns lifetime due to the meso-pores about 5 nm size [3], with intensity 24% for pure gel, 15% for the mix and none for the zeolite. These lifetimes can be used as unique fingerprints for characterizing zeolite versus silica gels and their mixtures.

The presence of a lifetime over 40 ns in silica gel in air suggest that the silica macro-pores are either closed or interconnected through the nano-channels that cannot be easily penetrated by oxygen, a prominent Ps quenching agent [4]. The CDB analysis of ratio curves indicates discrete structure of pure materials compared to less-defined structure of the mixture caused by a possible hybridization of the atomic states.

Novel positron annihilation spectroscopy results presented here will allow greater in-depth studies of the process of catalyst deterioration and deactivation and to understand how the process conditions of the chemical reactor affect the internal structure of the catalyst (e.g. via calcination, coking). Since both materials have distinct characteristics indicated by the PAL fingerprints due to different nature of internal structures, we hope to be able to study the process conditions that most greatly impact material ageing.

References

- [1] J. Kansy. *Nuclear Instr. and Methods*, 374, is.2, p.235-244, (1996).
- [2] Y. C. Yean, *Principles of Applications of Positron and Positronium Chemistry*, ISBN-978-981-238-144-8, (2003).
- [3] T. Goworek, K. Ciesielski, and al, *Chemical Physics*, 230, pp. 305-315 (1998).
- [4] R. Zaleski, M. Sokol, *Materials Science Forum*, Vol.666, pp 123-128 (2011).

*Corresponding author, Email: jagoda.urban-klaehn@inl.gov