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Micro-engineered catalyst systems: ABB's advancement in structured catalytic packings

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Abstract

ABB has advanced catalysis with micro-engineered catalyst (MEC) systems by providing a uniquely small particle size on a formable catalyst support through the integration of catalysis and reaction engineering. A mechanically strong catalytic web of micro-fibers has been engineered and shaped utilizing both computational fluid dynamics (CFD) and cold flow experiments to optimize flow characteristics. This article discusses techniques used for the development of novel catalytic structured packings for catalytic distillation applications. CFD models (verified through experiments performed on small-sized structures) were shown to be of great utility in screening new structure ideas. Results will illustrate achievement of both high gas–liquid contacting and bulk mixing at low pressure drop with the potential to provide enhanced catalyst utilization by taking advantage of the intrinsic MEC properties, particularly its high porosity and exposed geometric fiber and catalyst surface area. This was shown by the successful testing of one of these catalyzed structures in the selective hydrogenation of C4 acetylenes. © 2001 Published by Elsevier Science B.V.

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1. Introduction

In recent years there has been much emphasis on "process intensification", i.e. reduction in the physical size and/or cost of a chemical plant at a given production rate [1]. In many processes the reaction section is the major size/cost component. This has placed focus on improving the integration of catalyst and reactor systems not only at the molecular level, but also by concentrating on the interaction between chemical reaction and heat- and mass-transfer properties.

Catalysts are usually made as millimeter-sized particles with the active catalytic agents dispersed on the surface of/or throughout the particle. Reactants then flow through a reactor containing the bulk-loaded catalyst. Arrangement, shape and size of the catalyst govern the flow dynamics and pressure drop. The flow pattern is random making fluid dynamics and heat-management difficult to predict and control. The effective use of active catalytic agents is sometimes inefficient due to the occurrence of internal mass-transfer limitations. These constraints result in higher raw material and operating costs as well as the production of less valuable by-products and waste.

ABB has invented a type of catalyst system that maximizes both internal and external catalyst mass-transfer while minimizing overall hydrodynamic

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constraints. Such "micro-engineered catalyst (MEC) systems" consist of a mechanically strong catalytic web of micro-fibers — thinner than a human hair — in which microscopically small catalyst particles are entrapped [2]. Fig. 1 shows two current MEC technology concepts. In both, a highly porous metal sheet is used as a rigid catalyst support for micron-sized catalyst particles. MEC systems achieve their advantage by effectively utilizing catalyst particles on a much smaller scale than currently applied in industry. The size of the catalyst particles used in an MEC is typically not greater than 50 μm, i.e. about two orders of magnitude size reduction, thereby greatly reducing internal mass-transfer limitations. The supports may be formed into structures tailored for a specific application, thereby optimizing the functions of catalysis and reaction engineering and thereby also limiting external mass-transfer limitations.

This paper describes the ongoing development of catalytic structured packings for applications requiring management of gas/liquid, liquid/solid and gas/solid mass-transfer by employing computational fluid dynamics (CFD) in concert with detailed hydrodynamic measurements. The development of structured MEC catalyst packings for two-phase applications is explained in detail. One example of such an application is catalytic distillation, which requires the integration of a chemical reaction and appropriate heat- and mass-transfer characteristics with the goal of maximizing separation and reaction efficiencies [3,4].

2. Structured packing design — current state-of-the-art

The generally accepted design criteria for non-catalytic structured distillation packings are:

- a large surface area for high interfacial contact,
- an open structure for low resistance to the flow (both phases),
- uniform liquid distribution across the surface,
- uniform vapor flow in the column cross-section [5,6].

Surface characteristics to improve spreading of the liquid on the surface have been found to be of critical importance for enhancing mass-transfer. Liquid spreading is effected via perforations and embossments, which also enhance mass-transfer due to turbulence in the liquid film improve "surface wetting" characteristics (low surface tension) of the packing material, and act to cause coalescence and break-up of liquid droplets [6–11]. It was further found that with porous materials, capillary effects have a negative effect on liquid hold-up [7,12–14]; however, liquid spreading and wetting, particularly at low liquid flows are enhanced due to the capillary forces [12].

To achieve good mixing the flow should be three-dimensional, encompass a high turbulence intensity, have a high mean velocity gradient, and exhibit a frequent change of direction of the main streams [15,16]. High shear is desired, and created by flow streams of different orientations crossing or touching each other. A preferred packing geometry should spread the flow in a three-dimensional manner and should not have predominant characteristics in specific directions.

3. Experimental

Initially, a series of experimental techniques was applied to benchmark existing commercial packings in single- and two-phase environments. The self-made packings were manufactured from various materials, viz. metal sheet, gauze (wire diameter 0.2 mm, opening 1.0 mm), and a 90% void, 12 μm micro-fiber felt used to manufacture the MEC structure.

In single-phase flow, laser doppler anemometry (LDA) was applied to measure velocity gradients and laser induced fluorescence (LIF) to measure mixing [17–22].

3.1. CFD modeling

To develop new packing geometries, single-phase flow CFD modeling tools (Fluent UNS with unstructured gridding) were used and the results compared and validated against experimental data from cold-flow modeling.

Initially, a CFD grid was set up to model as closely as possible to the small experimental test rig. The rig inlet was configured to reduce any velocity non- uniformities, and consisted of a channel 30 mm in both height and width, which contained a three layer section of the packing. The overall length was 60 mm.

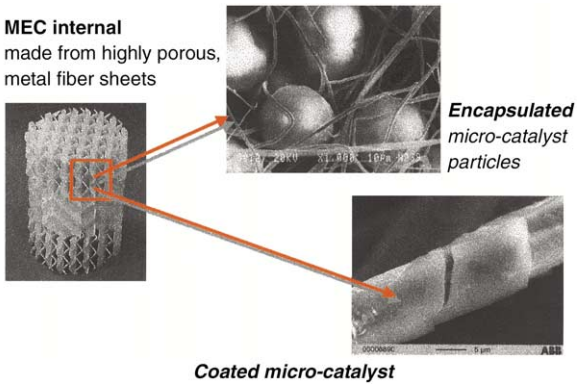

Fig. 1. MEC system concepts.

LDA measurements were made on planes close to the exit of a common corrugated structured packing using water at an F-factor (defined as $u\rho^{1/2}$ and measured in SI units) of around 130.

The CFD grids created were tetrahedral with around 300,000 cells. A top-hat velocity profile was assumed at the inlet, which was positioned 10 mm in front of the packing, and the domain outlet was 30 mm downstream of the packing exit plane. This allowed enough space to measure and compare two planes. Two grids of this kind were created, one without any perforations in the packing sheet, and the other with circular holes 4 mm in diameter. The holes in the computational grid were positioned identically to those in the experimental rig.

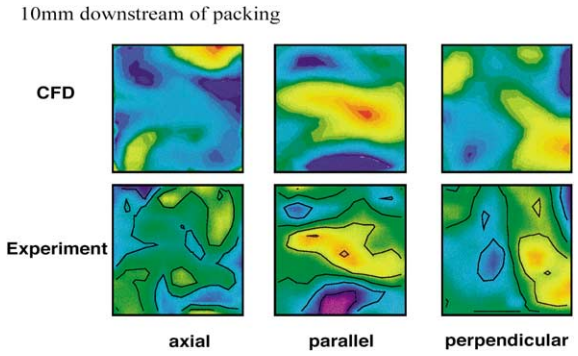

Fig. 3. Comparison between actual CFM experiment and CFD-simulated experiment of normalized velocity distributions for a corrugated structured sheet packing with holes.

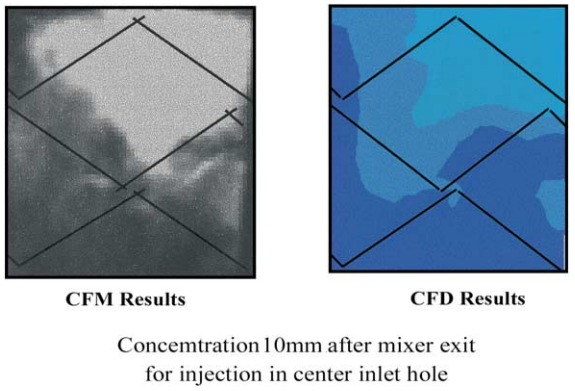

Fig. 4. Comparison between actual CFM experiment and CFD-simulated experiment for the dye distribution downstream of a corrugated structured sheet packing with holes.

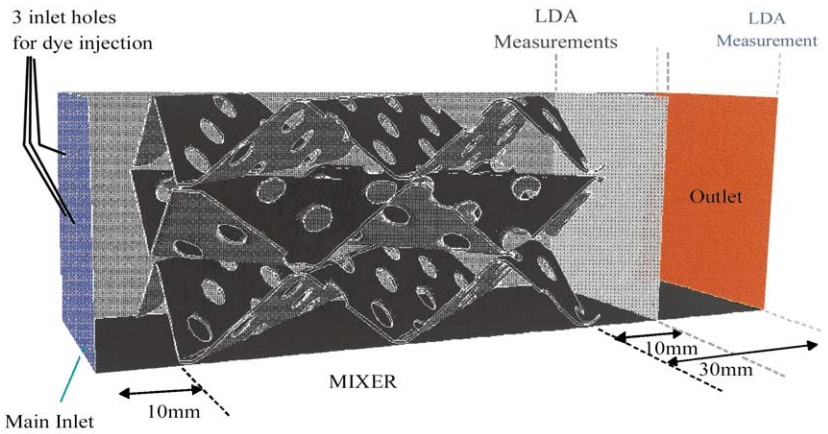

Fig. 2. Experimental approach to quantify mixing performance of packings.

3.2. Cold flow testing

For calibration tests, miniature corrugated packing geometries were assembled. To determine the effect of packing material on fluid dynamic performance, structures were made from both sheet metal containing small (4 mm diameter) holes, and also from a 12 μm micro-fiber felt. These miniature geometries were measured using the experimental approach presented in Fig. 2. Results of cold flow modeling (CFM) measurements expressed as a normalized velocity distribution in various directions were determined for water at an *F*-factor of ca. 130. Measurements were taken 10 and 30 mm downstream of the packing. Fig. 3 shows the quantitative normalized comparison between experimental CFM and the CFD simulations. Represented are the results at 10 mm downstream of the positions downstream of the corrugated structured packing made of a solid foil with 4 mm circular holes. Comparing the data from both the CFM and CFD results at a position 10 as well as 30 mm downstream of the packing clearly shows that the CFD simulation accurately predicted the experimental result. This is further demonstrated by comparing an actual camera picture with a CFD-generated picture illustrating mixing capability represented by injected dye distribution (Fig. 4). A similar result was also seen with two other benchmarked structures: structured packings made of a solid foil and felt material without punched holes. Oxygen desorption from water was used to measure gas–liquid mass-transfer rates.

4. Results and discussion

4.1. Characterization of standard structured packings: velocity and liquid distributions

Fig. 5 shows a comparison between a Sulzer Mellapak® 250 Y and a self-made packing from a 90% void, 12 μm micro-fiber felt with a similar geometry, but without holes or special surface characteristics. Both grossly show similar undulations in the velocity ratios indicating that the felt has similar mixing/flow distribution characteristics as the solid sheet with holes. Even though the felt is 90% porous, it is quite likely that filling of the relatively narrow pores (around 30 μm average) with liquid results in a solid barrier to bulk flow.

The ability to distribute liquid was also measured. Even though capillary forces may result in an improved capacity, they are known to have a negative effect on liquid hold-up [7,13,14,26]. However, liquid spreading and wetting, particularly at low liquid flows should be enhanced.

Sulzer Optiflow® packing provides excellent liquid distribution compared to a Mellapak. We believe that this is based on a number of positive design features in the Optiflow packing:

- a large number of drip points,
- frequent liquid film renewal,
- the ability to properly disperse the liquid flow.

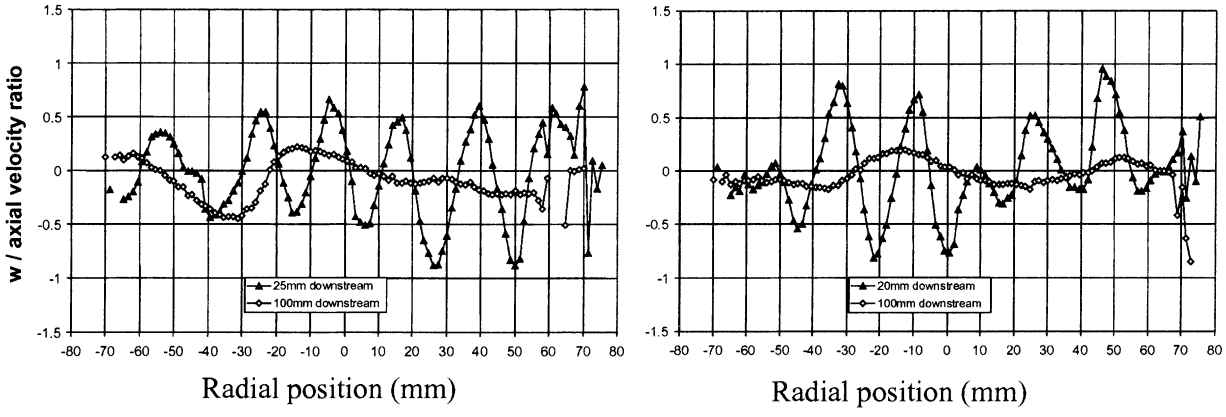

Fig. 5. Results of LDA measurements. Left: Sulzer Mellapak® 250 Y; right: self-assembled packing (250 m^2/m^3) made of 90% void micro-fiber felt.

Thus, the performance of a corrugated structure can be improved by:

- openings to allow gas distribution perpendicular to the sheets,
- a large number of connecting points,
- adding drip points to transport liquid from one surface to the other,
- good contact between adjacent sheets for improved liquid distribution.

In the design of MEC structures, it was decided that the above-mentioned features should be combined with the other design criteria for structured packings. The Optiflow design is not intended as a mixer, but rather as a low pressure drop distillation packing (lack of large scale vortices, low p [27]). This structure also has a relatively low geometric surface area, and thus may not be a good packing design for MEC applications, which require geometric surface area to allow volumetric catalytic activity.

4.2. CFD modeling to design novel MEC structured packings

Novel packing configurations were modeled to determine appropriate packing characteristics for improved mixing at low pressure drop. The tested computational geometries were all periodic and, therefore, a relatively small computational domain could model an infinitely large packing structure. Each grid was again generated using a tetrahedral mesh with each domain containing approximately 50,000 cells.

Several novel packing designs were evaluated in the developed, calibrated, single-phase CFD model. The most desirable designs were selected on the basis of large turbulent kinetic energy and high pressure drop of the structure to enhance both mass-transfer through the fiber felt (so called "cross-flow") and forced flow into the structure. However, the overall pressure drop over the length of the packing had to remain tolerable [5,7–9,23–25]. The goal was to enhance bulk mass-transfer and to increase catalyst utilization throughout the thickness of the porous medium.

We "tested" the novel structure concepts using the previously mentioned benchmark tool. Two generic design concepts (shown in Fig. 6) that yielded positive results from the CFD benchmarking tool were selected on the basis of manufacturability and desired performance criteria. Both showed a large turbulent kinetic energy and high pressure drop across the faces of the structure at acceptable overall pressure drop over the length of the packing. CFD results for these specific features are shown in Fig. 7. The calculation showed that the intrinsic pressure drop of the vertical channel design is lower than any of the corrugated designs at equivalent geometric surface area. The actual pressure drop of the vertical channel design at $280\,m^2/m^3$ surface area is about equal to the standard corrugated packing design (with holes) with less surface area ($250\,m^2/m^3$). The addition of the special surface features (vortex generators) results in only a minimal increase in pressure drop. Vortex-generating punch-outs allow the addition of holes at no expense of geometric sheet surface area, which is then available for the catalytic reaction. The sharp pointing edges of the punch-outs will enhance continuous liquid renewal and the increased localized eddies should act to reduce flow channeling [23,24]. The punch-outs can be modified to produce sufficient channel obstruction to minimize the occurrence of this undesirable channeling. Multiple geometrical modifications are thus possible [28,29].

Finally, the new MEC structure designs were manufactured from micro-fiber felt with specially designed tools [30]. Mass-transfer rates were measured using oxygen desorption from water in two operating regimes of low and high liquid flow rate in a countercurrent air stream. Fig. 8 shows the results in the case of low flow rate, showing how comparable gas–liquid mass-transfer rates and overall performance are obtained for the porous structures (corrugations, vertical channel) and a commercially available Mellapak® 250 Y structure. This is remarkable because the frontal blockage of the MEC packings is about five times larger than that of the Mellapak structure due to the differences in sheet thickness. Also, the characteristics of the micro-fibrous sheet, such as capillary forces, would be expected to attribute to intrinsically poorer performance. This would suggest that the novel designs — if constructed from a flat metal sheet — would have superior mass-transfer characteristics.

At higher liquid flow rates, the Mellapak® structure has better oxygen desorption rates than the vertical channel design; however, the capacity range of the vertical channel design seems to be larger [26]. At high flow rate, the highest desorption rates are

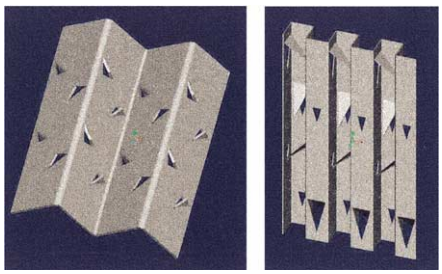

Fig. 6. Two new families of structured packing designs. Left: corrugated packing design with surface vortex generators; right: vertical channel design.

measured with the corrugated structure with vortex generators. We ascribe this to the observed bubble formation at the vortex generator points, which increase the mass-transfer area and enhance gas–liquid mass-transfer [5,9]. In addition to this positive feature, the capacity of this packing seems to be enhanced, as evidenced by it's drop-off in performance occurring at a higher gas rate.

One of these catalyzed structures was successfully tested in the selective hydrogenation of C4 acetylenes, wherein it was proven that both catalyst utilization and hydrodynamic characteristics could be improved. Due to their proprietary nature, detailed results cannot be included herein. However, Fig. 9 shows how the

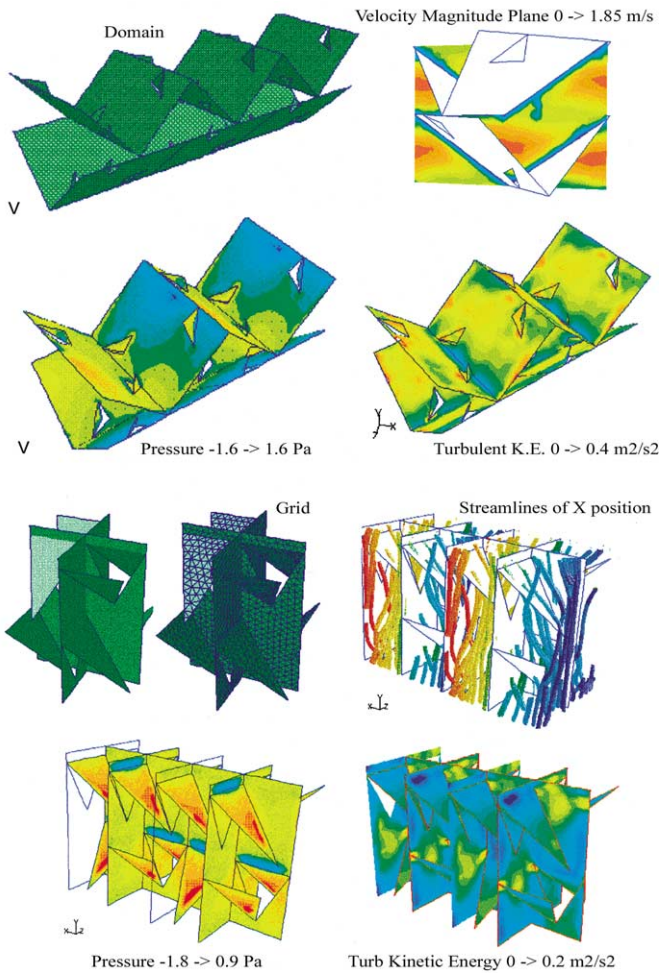

Fig. 7. CFD results for the two new families structured packing designs; increased darkening in color indicates larger differential in values.

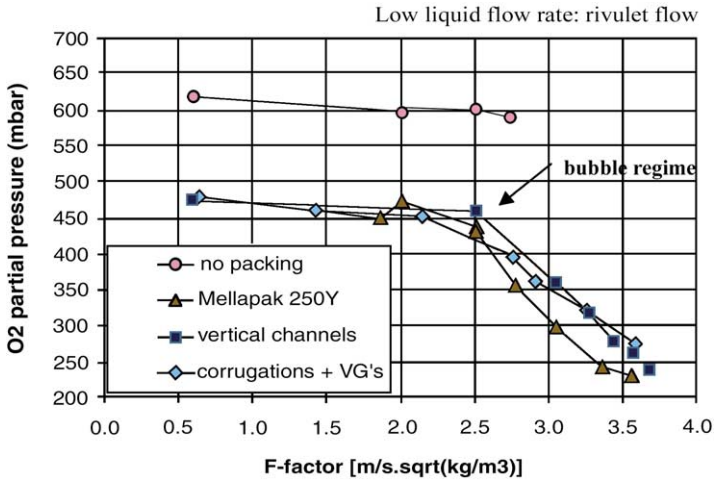

Fig. 8. Mass-transfer rate comparison, measured using oxygen desorption from water at low liquid rate, between new MEC packing designs and a Sulzer Mellapak® 250 Y packing.

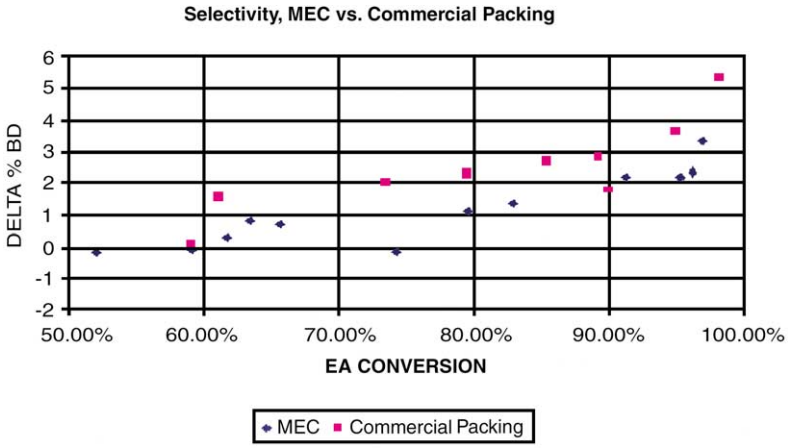

Fig. 9. MEC concept applied to catalytic distillation (selective hydrogenation of C4 acetylenes).

selectivity of the triple bound hydrogenation compared to double bond hydrogenation has been improved by careful selection of catalyst activity and hydrodynamic structure.

5. Conclusions

Novel structured catalyst packings have been designed based on the use of verified computational fluid dynamics and selected cold flow experiments. These packing designs take advantage of the unique properties of a new type of catalyst support material: a highly porous micro-fiber felt consisting of small metal fibers. By being more than 90% porous, the felt offers significantly greater exposed catalyst surface area and, therefore, enhanced catalyst utilization. Using this approach, separately optimized bulk transport properties can be combined with enhanced catalyst utilization.

CFD allowed both the determination of overall bulk parameters (such as pressure drop and mixing) and local parameters (such as local pressure drop across

the MEC fiber felt). It was used to screen a number of novel structural designs without performing expensive, time-consuming experimental tests.

Two families of structural designs were found in this study: a corrugated packing design with surface vortex generators and a vertical channel design. Both the designs can be manufactured with specially designed tools without compromising the nature of the micro-fiber felt substrate. Two-phase mass-transfer testing was performed on the new designs, which were compared with a commercially available product (Sulzer Mellapak® 250 Y). In these tests at low liquid flow rate, comparable gas–liquid mass-transfer rates and overall performance were obtained for the new packing designs made of the porous MEC material. This is remarkable, since the frontal blockage of the MEC packings is about five times larger than that of the Mellapak structure and certain characteristics of the micro-fibrous sheet, such as capillary forces, would be expected to be attributed to intrinsically poorer performance.

By introducing a novel catalyst support material — the high surface area micro-fiber felt — optimal bulk transport can be combined with enhanced catalyst utilization.

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